

## DECLARATION

I, Katsuramaru Fujita, a national of Japan, c/o Sumitomo Chemical Intellectual Property Service, Limited, 5-33, Kitahama 4-chome, Chuo-ku, Osaka 541-8550, Japan, declare that to the best of my knowledge and belief the attached is a full, true, and faithful translation into English made by me of the priority document, Japanese Patent Application No. 2002-86173.

Signed this 11th day of April, 2008

Katsuramaru Fujita

English Translation of JP application No. 2002-086173

[Name of Document] PATENT APPLICATION  
[Docket No.] P154187  
[Filing Date] March 26, 2002  
[Addressee] Commissioner, Patent Office  
[International Patent Classification]  
H05B 33/14  
C07F 15/00  
[Inventor]  
[Address] 6, Kitahara, Tsukuba-shi, Ibaraki  
c/o Sumitomo Chemical Company, limited  
[Name] Satoshi KOBAYASHI  
[Inventor]  
[Address] 6, Kitahara, Tsukuba-shi, Ibaraki  
c/o Sumitomo Chemical Company, limited  
[Name] Shuji DOI  
[Applicant]  
[Identification No.] 000002093  
[Name] Sumitomo Chemical Company, limited  
[Agent]  
[Identification No.] 100093285  
[Patent Attorney]  
[Name] Takashi KUBOYAMA  
[Telephone No.] 06-6220-3405  
[Elected Agent]  
[Identification No.] 100094477  
[Patent Attorney]  
[Name] Naoyoshi JINNO  
[Telephone No.] 06-6220-3405  
[Elected Agent]  
[Identification No.] 100113000  
[Patent Attorney]  
[Name] Tohru NAKAYAMA  
[Telephone No.] 06-6220-3405  
[Elected Agent]  
[Identification No.] 100119471  
[Patent Attorney]

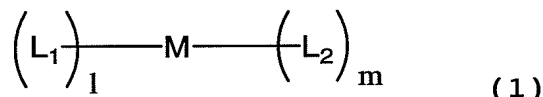
[Name] Masayuki ENOMOTO  
[Telephone No.] 06-6220-3405  
[Indication on Fees]  
[Prepayment Book No.] 010238  
[Amount of Payment] 21,000 yen  
[List of Articles Filed]  
[Name of Article] Specification 1  
[Name of Article] Abstract 1  
[Number of General Power] 0109029  
[Necessity of Proof] Yes

[Name of Document] SPECIFICATION

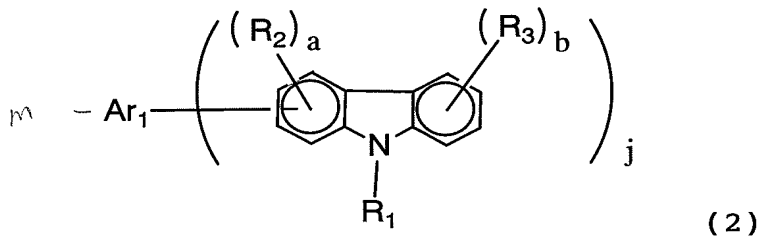
[Title of the Invention] METAL COMPLEX, POLYMER COMPOUND AND ORGANIC ELECTROLUMINESCENT DEVICE

[Scope of Claims for Patent]

1. A complex represented by the below formula (1), and having phosphorescence in a visible region,

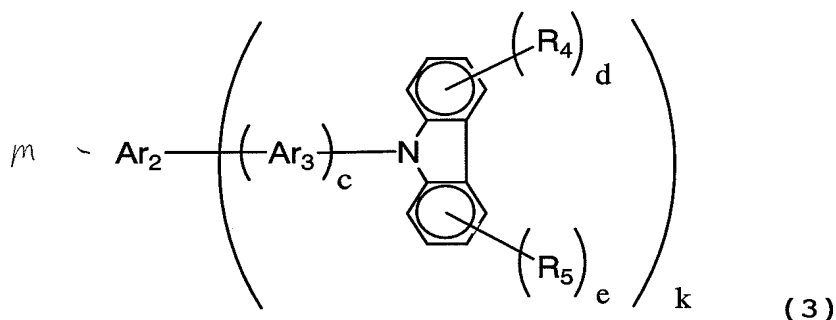


(wherein, M is a metal which is an atom having an atomic number of 50 or more, and intersystem crossing between a singlet state and a triplet state can occur in this complex by spin-orbit interaction.  $L_1$  represents a ligand represented by the following formula (2) or formula (3).  $L_2$  represents: a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom; a halogen atom; or a hydrogen atom.  $l$  represents an integer of 1-3.  $m$  represents an integer of 0-3. When  $m$  is two or more,  $L_2$ s may be the same or different.  $l+m$  is an integer of 2-5.)



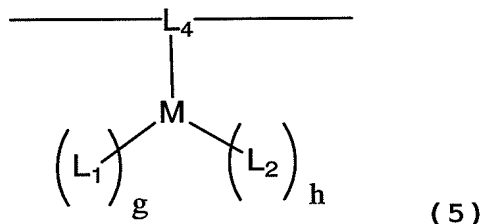
(wherein,  $Ar_1$  represents a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has  $j$  pieces of substituents having carbazole ring structure.  $j$  represents an integer of 1 to 3.  $R_2$  to  $R_3$  each independently represent a halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyloxy group, imino group, amide

group, arylalkenyl group, arylalkynyl group, cyano group, or a monovalent heterocyclic group.  $R_1$  represents alkyl group, aryl group, arylalkyl group, arylalkenyl group, arylalkynyl group, or a monovalent heterocyclic group.  $j$  represents an integer of 1-3.  $a$  represents an integer of 0 to 3.  $b$  represents an integer of 0 to 4. When  $a$  or  $b$  is two or more, each  $R_2$  or  $R_3$  may be mutually same or different, and may be connected to form a ring.)

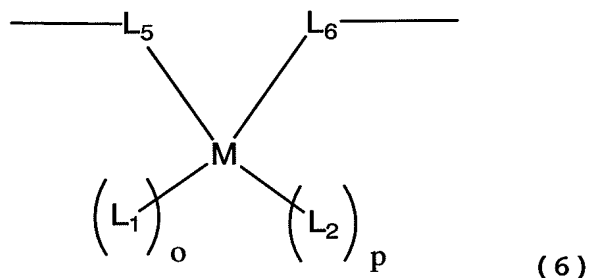


(wherein,  $\text{Ar}_2$  represents a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to  $k$  pieces of  $\text{Ar}_2$ s.  $k$  represents an integer of 1-3.  $\text{Ar}_3$  represents a group selected from an alkylene group, arylene group and divalent heterocyclic group.  $R_4$  and  $R_5$  each independently represent a halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or a monovalent heterocyclic group.  $k$  represents an integer of 1 to 3.  $c$  represents 0 or 1.  $d$  and  $e$  each independently represent an integer of 0 to 4. When  $d$  or  $e$  is two or more, each  $R_4$  or  $R_5$  may be mutually same or different, and may be connected to form a ring.)

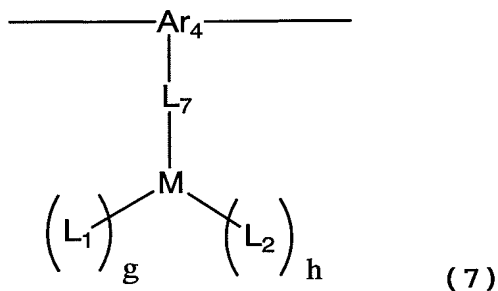
2. A polymeric compound comprising a repeating unit represented by the below formula (5), (6) or (7), which has phosphorescence in a visible region,



(wherein, M, L<sub>1</sub>, and L<sub>2</sub> are the same as those of formula (1) of Claim 1. L<sub>4</sub> is a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to two neighboring repeating units with two connecting bonds. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub>s may be mutually same or different. g+h is an integer of 1-4.)

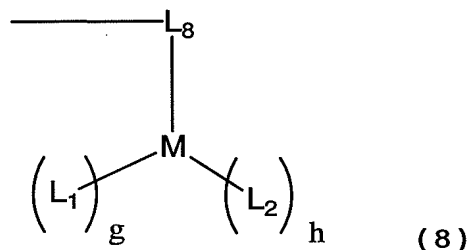


(wherein, M, L<sub>1</sub>, and L<sub>2</sub> are respectively the same as those of Formula (1) in Claim 1. L<sub>5</sub> and L<sub>6</sub> are each independently, a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to neighboring repeating unit with two connecting bonds, respectively. o represents an integer of 1-3 and p represents an integer of 0-3. When p is two or more, L<sub>2</sub>s may be the same or different. o+p is an integer of 1-3.)



(wherein, M, L<sub>1</sub>, and L<sub>2</sub> are respectively the same as those of formula (1) in Claim 1. Ar<sub>4</sub> is an arylene group or a divalent heterocyclic group. L<sub>7</sub> is a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has a covalent bond to Ar<sub>4</sub> with one connecting bond. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub>s may be the same or different. g+h is an integer of 1-4.).

3. A polymer compound having a structure represented by the below formula (8) and has phosphorescence in a visible region.



(wherein, M, L<sub>1</sub>, L<sub>2</sub> are respectively the same as those of formula (1) in Claim 1. L<sub>8</sub> is a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has a covalent bond at the polymer terminal with one connecting bond. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub> may be the same or different. g+h is an integer of 1-4.)

4. An organic electroluminescence device comprising a layer which contains the complex of Claim 1 or the polymer compound of Claim 2 or 3, between electrodes consisting of an anode and a cathode.

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

The present invention relates to a new complex and a new polymer compound, manufacture method thereof and a light-emitting device using the metal organic complex, or the polymer compound as a luminescence material.

[0002]

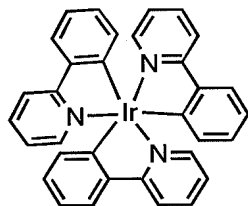
[Prior Art]

As the light-emitting material having phosphorescence in visible region used for a light emitting layer of light-emitting device, devices using a metal complex (hereafter may be referred to as triplet light-emitting complex) showing light emission from triplet excited state have been known.

[0003]

As the triplet light-emitting complex, for example, Ir(ppy)<sub>3</sub> which includes iridium as the central metal, (Appl.Phys.Lett.,75,4(1999)), PtOEP which includes platinum as the central metal (Nature,395,151(1998)), Eu(TTA)<sub>3</sub>phen which includes europium as the central metal (Jpn. J. Appl. Phys.,34,1883(1995)) are known.

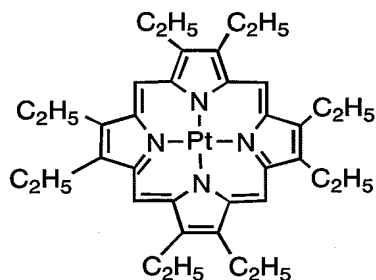
[0004]



Ir(ppy)<sub>3</sub>

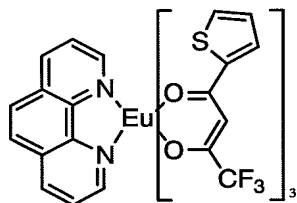
[0005]





PtOEP

[0006]



Eu(TTA)<sub>3</sub>phen

[0007]

[Problems to be Solved by the Invention]

However, the light emitting efficiency of the device using the above-mentioned well-known complex was not yet satisfactory.

The object of the present invention is to provide a complex and a polymer compound which are excellent in light emitting efficiency, and have phosphorescence in visible region,.

[0008]

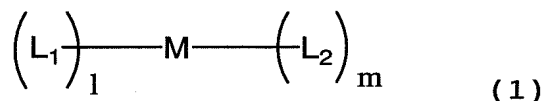
[Means for Solving the Problems]

As a result of intensive studies to solve the above-mentioned problem, the present inventors found out that the complex of following (1) having a ligand having a specific substituent derived from a carbazole ring, and the polymer compound of following (5) - (8), have phosphorescence in visible region, and have high light emitting efficiency, and have accomplished the present invention.

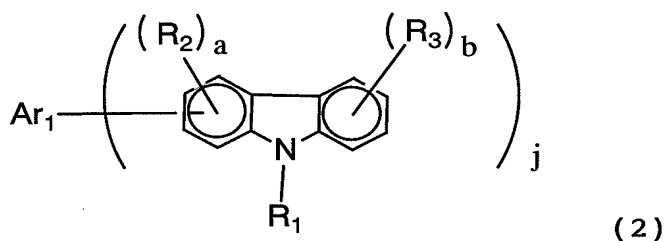
[0009]

That is, the present invention relates to the following [1]- [3].

[1] A complex represented by the below formula (1), and having phosphorescence in a visible region,

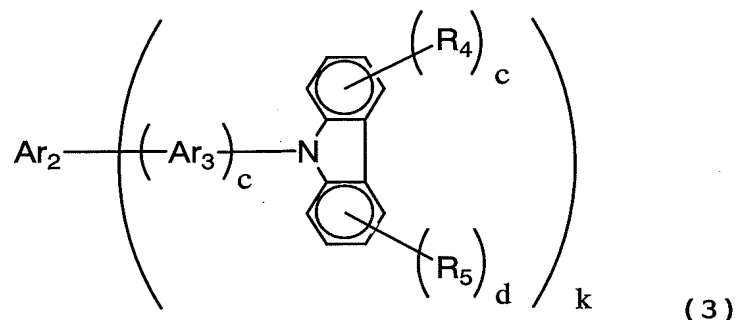


(wherein, M is a metal which is an atom having an atomic number of 50 or more, and intersystem crossing between a singlet state and a triplet state can occur in this complex by spin-orbit interaction.  $L_1$  represents a ligand represented by the following formula (2) or formula (3).  $L_2$  represents: a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom; a halogen atom; or a hydrogen atom. 1 represents an integer of 1-3. m represents an integer of 0-3. When m is two or more,  $L_2$ s may be the same or different. 1+m is an integer of 2-5.)



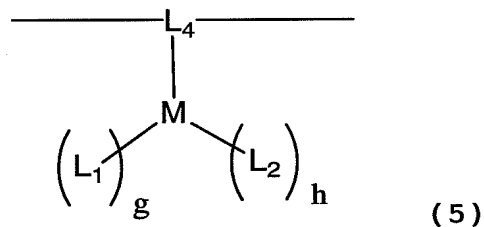
(wherein,  $Ar_1$  represents a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has j pieces of substituents having carbazole ring structure. j represents an integer of 1 to 3.  $R_2$  to  $R_3$  each independently represent a halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or a monovalent heterocyclic group.  $R_1$  represents alkyl group, aryl group, arylalkyl group, arylalkenyl group, arylalkynyl group, or a monovalent heterocyclic group. j represents an integer of 1-3. a represents an integer of 0 to 3. b represents an integer of 0 to 4. When a or b is two or more, each  $R_2$  or  $R_3$  may be mutually same or different, and may be connected to

form a ring.)

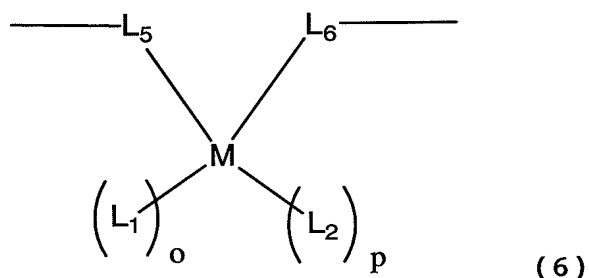


(wherein, Ar<sub>2</sub> represents a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to k pieces of Ar<sub>2</sub>s. k represents an integer of 1-3. Ar<sub>3</sub> represents a group selected from an alkylene group, arylene group and divalent heterocyclic group. R<sub>4</sub> and R<sub>5</sub> each independently represent a halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or a monovalent heterocyclic group. k represents an integer of 1 to 3. c represents 0 or 1. d and e each independently represent an integer of 0 to 4. When d or e is two or more, each R<sub>4</sub> or R<sub>5</sub> may be mutually same or different, and may be connected to form a ring.)

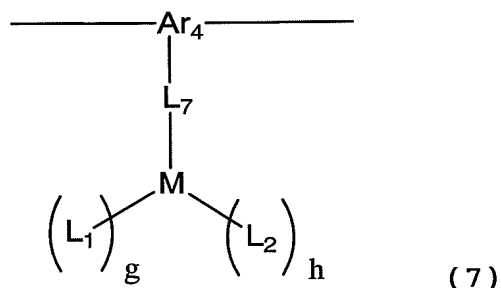
[2] A polymeric compound comprising a repeating unit represented by the below formula (5), (6) or (7), which has phosphorescence in a visible region,



(wherein, M, L<sub>1</sub>, and L<sub>2</sub> are the same as those of formula (1) of Claim 1. L<sub>4</sub> is a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to two neighboring repeating units with two connecting bonds. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub>s may be mutually same or different. g+h is an integer of 1-4.)



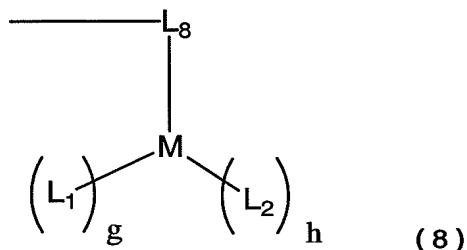
(wherein, M, L<sub>1</sub>, and L<sub>2</sub> are respectively the same as those of Formula (1) in Claim 1. L<sub>5</sub> and L<sub>6</sub> are each independently, a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to neighboring repeating unit with two connecting bonds, respectively. o represents an integer of 1-3 and p represents an integer of 0-3. When p is two or more, L<sub>2</sub>s may be the same or different. o+p is an integer of 1-3.)



(wherein, M, L<sub>1</sub>, and L<sub>2</sub> are respectively the same as those of formula (1) in Claim 1. Ar<sub>4</sub> is an arylene group or a divalent heterocyclic group. L<sub>7</sub> is a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has a covalent bond to Ar<sub>4</sub> with one connecting bond. g represents an integer of 1-3 and h represents

an integer of 0-3. When h is two or more, L<sub>2</sub>s may be the same or different. g+h is an integer of 1-4.).

[3] A polymeric having a structure represented by the below formula (8) and has phosphorescence in a visible region.

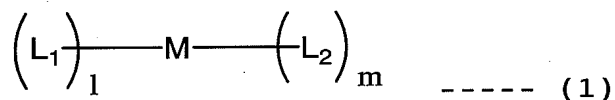


(wherein, M, L<sub>1</sub>, L<sub>2</sub> are respectively the same as those of formula (1) in Claim 1. L<sub>8</sub> is a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has a covalent bond at the polymer terminal with one connecting bond. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub> may be the same or different. g+h is an integer of 1-4.)

[0010]

[Modes for Carrying out the Invention]

The complex of the present invention is represented by the below formula (1), and is characterized by having phosphorescence in visible region.



[0011]

Here, M is a metal which is an atom having an atomic number of 50 or more, and intersystem crossing between a singlet state and a triplet state can occur in this complex by spin-orbit interaction.

[0012]

Examples of atoms represented by M include: a rhenium atom, osmium atom, iridium atom, platinum atom, gold atom, lanthanum atom, cerium atom, praseodymium atom, neodymium atom, promethium atom, samarium atom, europium atom, gadolinium atom, terbium atom, dysprosium atom, etc. Rhenium atom, osmium

atom, iridium atom, platinum atom, gold atom, samarium atom, europium atom, gadolinium atom, terbium atom, and dysprosium atom are preferable, and iridium atom, platinum atom, gold atom, and europium atom are more preferable.

[0013]

$L_2$  represents: a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom; a halogen atom; or a hydrogen atom.

[0014]

Examples of the ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, include an alkyl group, alkoxy group, carboxyl group, alkylthio group, alkyl amino group, aryl group, aryloxy group, arylthio group, arylamino group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkylamino group, sulfone oxy group, cyano group, a heterocyclic ligand, carbonyl ligand, ether ligand, phosphine ligand, phosphite ligand, and sulfide ligand.

[0015]

Examples of the alkyl group, alkoxy group, alkylthio group, alkylamino group, aryl group, aryloxy group, arylthio group, arylamino group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, and imino group, include the groups described in the following  $R_2$  and  $R_3$ .

[0016]

Examples of the heterocyclic ligands include heterocyclic ligands such as pyridine ring, pyrrole ring, thiophene ring, oxazole, and furan ring, and ligands consisting of connected benzene rings. Specifically, exemplified are: phenylpyridine, 2-(para-phenylphenyl)pyridine, 7-bromobenzo[h] quinoline, 2-(4-thiophene-2-yl)pyridine, 2-(4-phenylthiophene-2-yl)pyridine, 2-phenylbenzoxazole, 2-(para-phenylphenyl)benzoxazole, 2-phenylbenzothiazole, 2-(para-phenylphenyl)benzothiazole, 2-(benzothiophene-2-yl)pyridine, 1,10-phenanthroline, 2,3,7,8,12,13,17,18-octa ethyl-21H,23H-porphyrin, etc.

[0017]

The carboxyl groups have about 2 to 20 carbon atoms, and examples thereof include specifically, acetyl oxy group, trifluoroacetyl oxy group, propionyloxy group, and benzoyloxy group. Examples of the sulfoneoxy group include benzene sulfoneoxy group, p-toluene sulfone oxy group, methane sulfoneoxy group, ethane sulfoneoxy group, and trifluoromethane sulfoneoxy group.

[0018]

Examples of the carbonyl ligand include: ketones, such as carbon monoxide, and acetone, benzophenone; diketones, such as acetyl acetone, and acenaphthoquinone; and acetonates, such as acetylacetonate, dibenzomethylate, and thenoyltrifluoroacetate.

[0019]

Examples of the ether ligand include dimethyl ether, diethyl ether, tetrahydrofuran, 1,2-dimethoxy ethane, etc.

[0020]

Examples of the phosphine ligand include triphenyl phosphine, diphenylphosphinoethane, and diphenylphosphinopropane. Examples of the phosphite ligand include dimethylphosphite and diphenylphosphite.

[0021]

Examples of the sulfide ligand include dimethyl sulfide, diphenyl sulfide, and thioanisole.

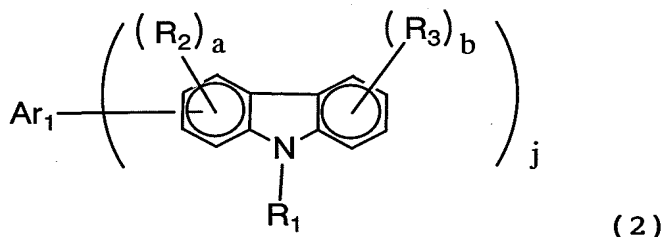
[0022]

l represents an integer of 1-3. m represent an integer of 0-3. When m is two or more,  $L_2$ s may be the same or different.  $l+m$  is an integer of 2-5.

[0023]

$L_1$  in formula (1) represents an ligand shown by the following formula (2) or formula (3).

[0024]



[0025]

Here,  $Ar_1$  represents a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has j pieces of substituents having carbazole ring structure. j represents an integer of 1 to 3.

[0026]

$R_2$  and  $R_3$  each independently represent a halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or a monovalent heterocyclic group.  $R_1$  represents alkyl group, aryl group, arylalkyl group, arylalkenyl group, arylalkynyl group, or a monovalent heterocyclic group.

[0027]

Examples of the halogen atom include fluorine, chlorine, bromine, and iodine.

[0028]

The alkyl group may be any of linear, branched or cyclic, and may have one or more substituents. The number of carbon atoms is usually from about 1 to 20, and specific examples thereof include methyl group, ethyl group, propyl group, i-propyl group, butyl group, i-butyl group, t-butyl group, pentyl group, cyclopentyl group, hexyl group, cyclohexyl group, heptyl group, octyl group, 2-ethylhexyl group, nonyl group, decyl group, 3,7-dimethyloctyl group, lauryl group, trifluoromethyl group, pentafluoroethyl group, perfluorobutyl group, perfluorohexyl group, perfluorooctyl



group, etc.; and pentyl group, hexyl group, octyl group, 2-ethylhexyl group, decyl group, and 3,7-dimethyloctyl group are preferable.

[0029]

The alkoxy group may be any of linear, branched or cyclic, and may have one or more substituents. The number of carbon atoms is usually from about 1 to 20, and specific examples thereof include methoxy group, ethoxy group, propyloxy group, i-propyloxy group, butoxy group, i-butoxy group, t-butoxy group, pentyloxy group, cyclopentyloxy group, hexyloxy group, cyclohexyloxy group, heptyloxy group, octyloxy group, 2-ethyl hexyloxy group, nonyloxy group, decyloxy group, 3,7-dimethyl octyloxy group, lauryloxy group, trifluoromethoxy group, pentafluoroethoxy group, perfluorobutoxy group, perfluoro hexyl group, perfluorooctyl group, methoxymethyloxy group, 2-methoxyethyloxy group, etc.; and pentyloxy group, hexyloxy group, octyloxy group, 2-ethylhexyloxy group, decyloxy group, and 3,7-dimethyloctyloxy group are preferable.

[0030]

The alkylthio group may be any of linear, branched or cyclic, and may have one or more substituents. The number of carbon atoms is usually from about 1 to 20, and specific examples thereof include methylthio group, ethylthio group, propylthio group, i-propylthio group, butylthio group, i-butylthio group, t-butylthio group, pentylthio group, cyclopentylthio group, hexylthio group, cyclohexylthio group, heptylthio group, octylthio group, 2-ethylhexylthio group, nonylthio group, decylthio group, 3,7-dimethyloctylthio group, laurylthio group, trifluoromethylthio group, etc.; and pentylthio group, hexylthio group, octylthio group, 2-ethylhexylthio group, decylthio group, and 3,7-dimethyloctylthio group are preferable.

[0031]

The alkylamino group may be any of linear, branched or cyclic, and may be monoalkylamino group or dialkylamino group. The number of carbon atoms is usually from about 1 to 40, and

specific examples thereof include methylamino group, dimethyl amino group, ethylamino group, diethylamino group, propyl amino group, dipropylamino group, i-propylamino group, diisopropylamino group, butylamino group, i-butylamino group, t-butylamino group, pentylamino group, cyclopentylamino group, hexylamino group, cyclohexylamino group, heptylamino group, octylamino group, 2-ethylhexylamino group, nonylamino group, decylamino group, 3,7-dimethyloctylamino group, laurylamino group, cyclopentylamino group, dicyclopentylamino group, cyclohexylamino group, dicyclohexylamino group, pyrrolidyl group, piperidyl group, ditrifluoromethylamino group, etc.; and pentylamino group, hexylamino group, octylamino group, 2-ethylhexylamino group, decylamino group, and 3,7-dimethyl octylamino group are preferable.

[0032]

The alkylsilyl group may be any of linear, branched or cyclic, and the number of carbon atoms is usually from about 1 to 60. Specific examples thereof include trimethylsilyl group, triethylsilyl group, tripropylsilyl group, tri-i-propylsilyl group, dimethyl-i-propylsilyl group, diethyl-i-propylsilyl group, t-butylsilyldimethylsilyl group, pentyldimethylsilyl group, hexyldimethylsilyl group, heptyldimethylsilyl group, octyldimethylsilyl group, 2-ethylhexyl-dimethylsilyl group, nonyldimethylsilyl group, decyldimethylsilyl group, 3,7-dimethyloctyl-dimethylsilyl group, lauryldimethylsilyl group etc.; and pentyldimethylsilyl group, hexyldimethyl silyl group, octyldimethylsilyl group, 2-ethylhexyl-dimethyl silyl group, decyldimethylsilyl group, and 3,7-dimethyloctyl dimethylsilyl group are preferable.

[0033]

The aryl group may have one or more substituents, and the number of carbon atoms is usually from about 3 to 60. Specific examples thereof include phenyl group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl group (C<sub>1</sub>-C<sub>12</sub> represents the number of carbon atoms 1-12. Hereafter the same.), C<sub>1</sub>-C<sub>12</sub> alkylphenyl group, 1-naphtyl group,

2-naphtyl group, pentafluorophenyl group, pyridyl group, pyridazinyl group, pyrimidyl group, pyrazyl group, triazyl group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl group, and C<sub>1</sub>-C<sub>12</sub> alkylphenyl group are preferable.

[0034]

The aryloxy group may have one or more substituents on the aromatic ring, and the number of carbon atoms is usually from about 3 to 60. Specific examples thereof include phenoxy group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenoxy group, C<sub>1</sub>-C<sub>12</sub> alkylphenoxy group, 1-naphtyloxy group, 2-naphtyloxy group, pentafluorophenyloxy group, pyridyloxy group, pyridazinyl group, pyrimidyloxy group, pyrazyl group, triazyloxy group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenoxy group, and C<sub>1</sub>-C<sub>12</sub> alkylphenoxy group are preferable.

[0035]

The arylthio group may have one or more substituents on the aromatic ring, and the number of carbon atoms is usually from about 3 to 60. Specific examples thereof include phenylthio group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenylthio group, C<sub>1</sub>-C<sub>12</sub> alkyl phenylthio group, 1-naphthylthio group, 2-naphthylthio group, pentafluorophenylthio group, pyridylthio group, pyridazinylthio group, pyrimidylthio group, pyrazylthio group, triazylthio group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenylthio group, and C<sub>1</sub>-C<sub>12</sub> alkylphenylthio group are preferable.

[0036]

The arylamino group may have one or more substituents on the aromatic ring, and the number of carbon atoms is usually from about 3 to 60. Specific examples thereof include phenylamino group, diphenylamino group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenylamino group, di(C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl) amino group, di(C<sub>1</sub>-C<sub>12</sub> alkylphenyl) amino group, 1-naphtylamino group, 2-naphtylamino group, pentafluorophenylamino group, pyridyl amino group, pyridazinylamino group, pyrimidylamino group, pyrazylamino group, triazylamino group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkylphenylamino group and di(C<sub>1</sub>-C<sub>12</sub> alkylphenyl) amino group are preferable.

[0037]

The arylsilyl group may have one or more substituents on the aromatic ring, and the number of carbon atoms is usually from about 3 to 60. Specific examples thereof include triphenyl silyl group, tri-p-xylylsilyl group, tribenzylsilyl group, diphenylmethylsilyl group, t-butyldiphenylsilyl group, dimethylphenylsilyl group, etc.

[0038]

The arylalkyl group may have one or more substituents, and the number of carbon atoms is usually from about 7 to 60. Specific examples thereof include phenyl-C<sub>1</sub>-C<sub>12</sub> alkyl group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkyl group, C<sub>1</sub>-C<sub>12</sub> alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkyl group, 1-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkyl group, 2-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkyl group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkyl group, and C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkyl group are preferable.

[0039]

The arylalkoxy group may have one or more substituents, and the number of carbon atoms is usually from about 7 to 60. Specific examples thereof include phenyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group, C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group, 1-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group, 2-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxy phenyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group, and C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkoxy group are preferable.

[0040]

The arylalkylthio group may have one or more substituents, and the number of carbon atoms is usually from about 7 to 60. Specific examples thereof include phenyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group, C<sub>1</sub>-C<sub>12</sub> alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group, 1-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group, 2-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group, and C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkylthio group are preferable.

[0041]

The arylalkylamino group has usually about 7 to 60 carbon atoms. Specific examples thereof include phenyl-C<sub>1</sub>-C<sub>12</sub> alkyl

amino group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkylamino group, C<sub>1</sub>-C<sub>12</sub> alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkylamino group, di(C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkyl) amino group, di(C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkyl) amino group, 1-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkylamino group, 2-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkylamino group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkylamino group, and di(C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkyl) amino group are preferable.

[0042]

The arylalkylsilyl group has usually about 7 to 60 carbon atoms. Specific examples thereof include phenyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group, C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group, C<sub>1</sub>-C<sub>12</sub> alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group, 1-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group, 2-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group, phenyl-C<sub>1</sub>-C<sub>12</sub> alkyl dimethylsilyl group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group, and C<sub>1</sub>-C<sub>12</sub> alkyl phenyl-C<sub>1</sub>-C<sub>12</sub> alkylsilyl group are preferable.

[0043]

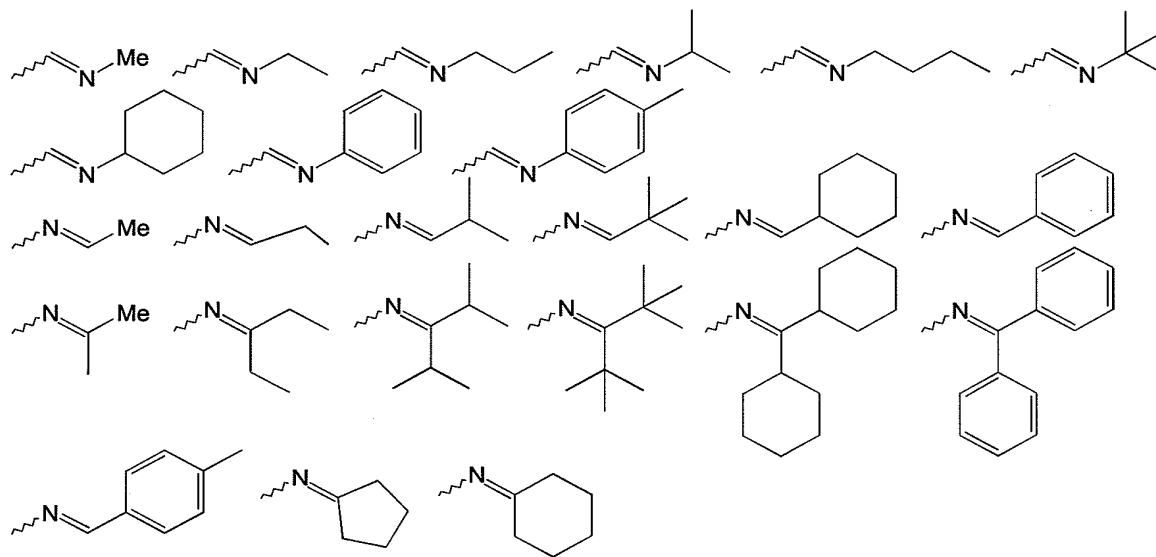
The acyl group has usually about 2 to 20 carbon atoms. Specific examples thereof include acetyl group, propionyl group, butyryl group, isobutyryl group, pivaloyl group, benzoyl group, trifluoroacetyl group, pentafluorobenzoyl group, etc.

[0044]

The acyloxy group has usually about 2 to 20 carbon atoms. Specific examples thereof include acetoxyl group, propionyloxy group, butyryloxy group, isobutyryloxy group, pivaloyloxy group, benzoyloxy group, trifluoroacetyloxy group, pentafluorobenzoyloxy group, etc.

[0045]

The imino group has usually about 2 to 20 carbon atoms. Specific examples thereof include the compounds represented by following formulas.



[0046]

The amide group has usually about 2 to 20 carbon atoms. Specific examples thereof include formamide group, acetamide group, propioamide group, butyroamide group, benzamide group, trifluoroacetamide group, pentafluorobenzamide group, diformamide group, diacetamide group, dipropioamide group, dibutyroamide group, dibenzamide group, ditrifluoroacetamide group, dipentafluorobenzamide group, etc.; and imides such as succinimide group and phthalic acid imide group, are also included.

[0047]

The arylalkenyl group has usually about 7 to 60 carbon atoms. Specific examples thereof include phenyl- $C_1$ - $C_{12}$  alkenyl group,  $C_1$ - $C_{12}$  alkoxyphenyl- $C_1$ - $C_{12}$  alkenyl group,  $C_1$ - $C_{12}$  alkylphenyl- $C_1$ - $C_{12}$  alkenyl group, 1-naphtyl- $C_1$ - $C_{12}$  alkenyl group, 2-naphtyl- $C_1$ - $C_{12}$  alkenyl group, etc.; and  $C_1$ - $C_{12}$  alkoxyphenyl- $C_1$ - $C_{12}$  alkenyl group, and  $C_1$ - $C_{12}$  alkylphenyl- $C_1$ - $C_{12}$  alkenyl group are preferable.

[0048]

The arylalkynyl group has usually about 7 to 60 carbon atoms. Specific examples thereof include phenyl- $C_1$ - $C_{12}$  alkynyl group,  $C_1$ - $C_{12}$  alkoxyphenyl- $C_1$ - $C_{12}$  alkynyl group,  $C_1$ - $C_{12}$

alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkynyl group, 1-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkynyl group, 2-naphtyl-C<sub>1</sub>-C<sub>12</sub> alkynyl group, etc.; and C<sub>1</sub>-C<sub>12</sub> alkoxyphenyl-C<sub>1</sub>-C<sub>12</sub> alkynyl group, and C<sub>1</sub>-C<sub>12</sub> alkylphenyl-C<sub>1</sub>-C<sub>12</sub> alkynyl group are preferable.

[0049]

The monovalent heterocyclic group means an atomic group in which a hydrogen atom is removed from a heterocyclic compound, and usually has about 4 to 60 carbon atoms. Specific examples thereof include thienyl group, C<sub>1</sub>-C<sub>12</sub> alkylthienyl group, pyroryl group, furyl group, pyridyl group, C<sub>1</sub>-C<sub>12</sub> alkylpyridyl group, etc.; and thienyl group, C<sub>1</sub>-C<sub>12</sub> alkylthienyl group, pyridyl group, and C<sub>1</sub>-C<sub>12</sub> alkylpyridyl group, are preferable.

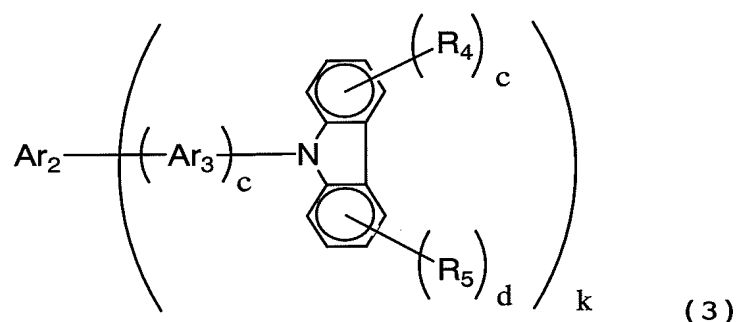
[0050]

R<sub>1</sub> represents an alkyl group, aryl group, arylalkyl group, aryl alkenyl group, arylalkynyl group, or a monovalent heterocyclic group. Specifically, the groups described in the above R<sub>2</sub> and R<sub>3</sub>.

[0051]

a represents an integer of 0-3. b represents an integer of 0-4. When a or b is two or more, each of R<sub>2</sub> and R<sub>3</sub> may be the same or different, and they are connected mutually to form a ring.

[0052]



[0053]

Here, Ar<sub>2</sub> represents a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to k pieces of Ar<sub>2</sub>s. k represents an integer of 1-3.

[0054]

The ligand represented by  $Ar_2$  is a ligand shown by  $L_2$  and has a bond with  $Ar_3$  at an arbitrary position.

[0055]

$Ar_3$  represents a group selected from an alkylene group, arylene group and divalent heterocyclic group.

In the group shown by  $Ar_3$ , as the alkylene group, methylene group, ethylene group, propylene group, cis-ethylenediyl group, trans-ethylenediyl group, and acetylenediyl group are exemplified. As the arylene group, phenylene group, naphthylene group, biphenylene group, anthracenediyl group, terphenylene group, pyrenediyl group, o-phenylene-vinylene group, m-phenylene-vinylene group, and p-phenylene-vinylene group are exemplified.  $c$  is 0 or 1.

[0056]

The divalent heterocyclic group means an atomic group in which two hydrogen atoms are removed from a heterocyclic compound, and the number of carbon atoms is about 4 to 20. As the heterocyclic compound here, furan, thiophene, pyrrole, pyrazole, pyridine, pyridazine, pyrimidine, pyradine, and bipyridyl are specifically exemplified

[0057]

$R_4$  and  $R_5$  each independently represent a halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or a monovalent heterocyclic group.  $k$  represents an integer of 1 to 3.  $c$  represents 0 or 1.  $d$  and  $e$  each independently represent an integer of 0 to 4. When  $d$  or  $e$  is two or more, each  $R_4$  or  $R_5$  may be mutually same or different, and may be connected to form a ring.

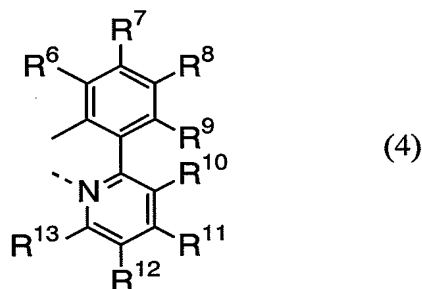
[0058]

The case where  $M$  of  $L_1$  bonds with at least one carbon atom



is preferable, and the case where it is a bidentate ligand which bonds with M and forms a 5 membered ring is more preferable.  
[0059]

More preferably, it is represented by the below formula (4).

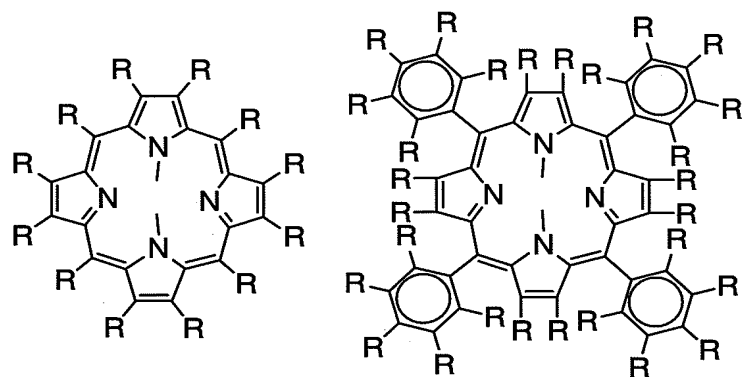
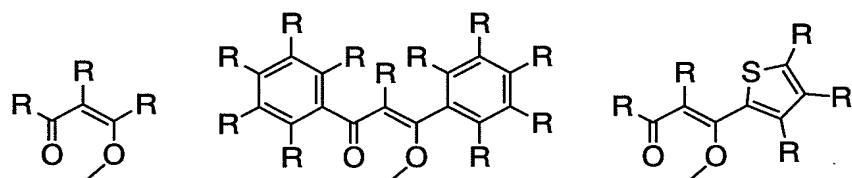


(In the formula,  $R^6$  to  $R^{13}$  each independently represent a hydrogen atom, halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkylamino group, arylalkylsilyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, monovalent heterocyclic group.  $R^6$  to  $R^{13}$  may be connected to form a ring. At least one of  $R^6$  to  $R^{13}$  is a carbazole group represented by (2) or a connecting group with  $Ar_3$ .)

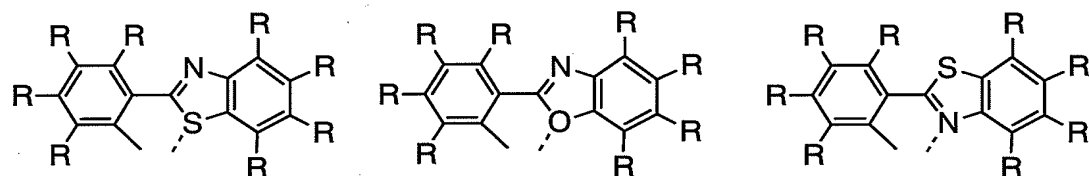
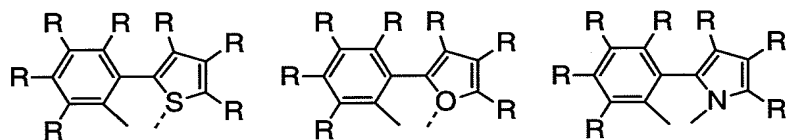
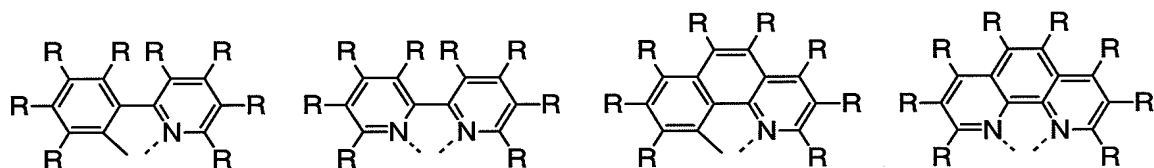
[0060]

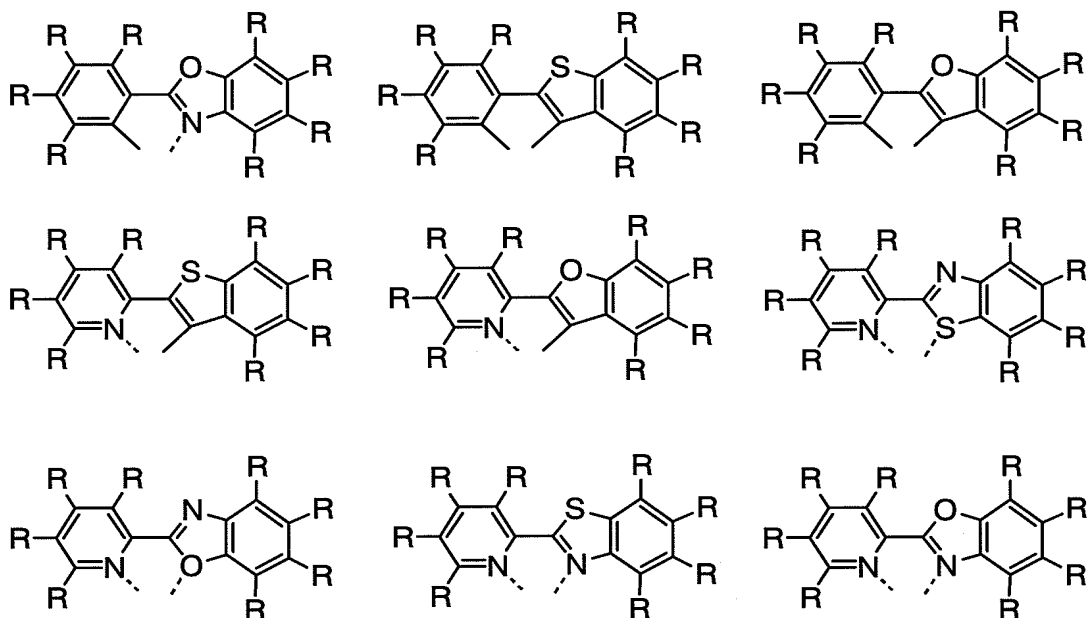
Examples of  $L_1$  include followings.

[0061]



[0062]





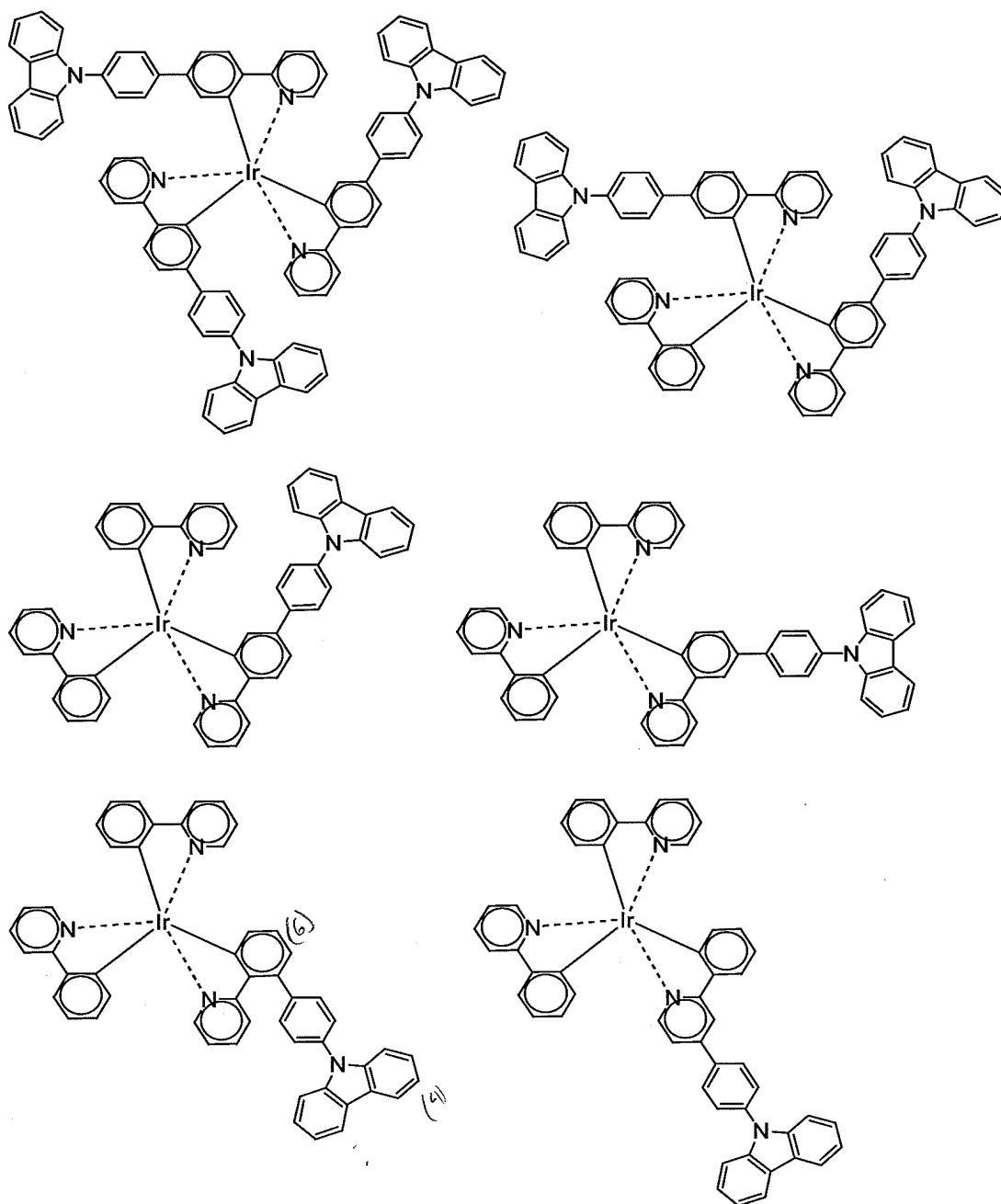
[0063]

Here, R each independently represent a hydrogen atom, halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, aryl alkoxy group, arylalkylthio group, arylalkylamino group, arylalkylsilyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, OR monovalent heterocyclic group. Specific examples include those represented in the above R<sub>2</sub> and R<sub>3</sub>. In each ligand, at least one R is a carbazole ring or a connecting group with Ar<sub>3</sub>. Rs may be connected mutually to form a ring. In order to improve the solubility into a solvent, it is preferable that at least one of the R has a long alkyl group.

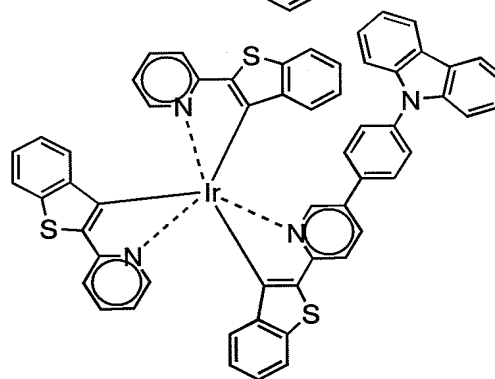
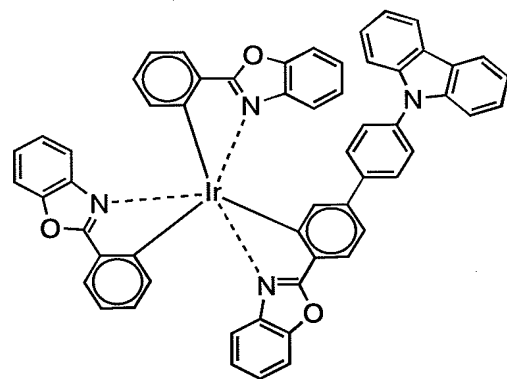
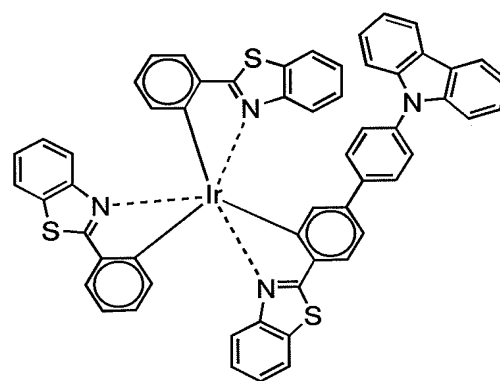
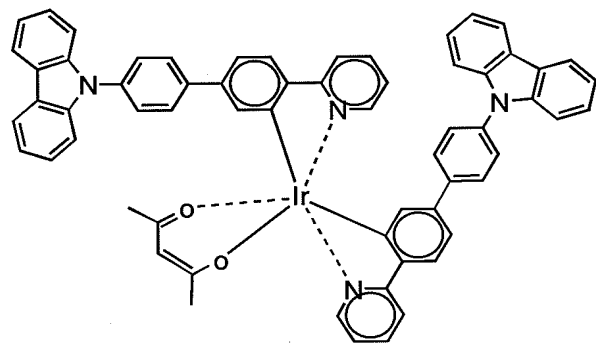
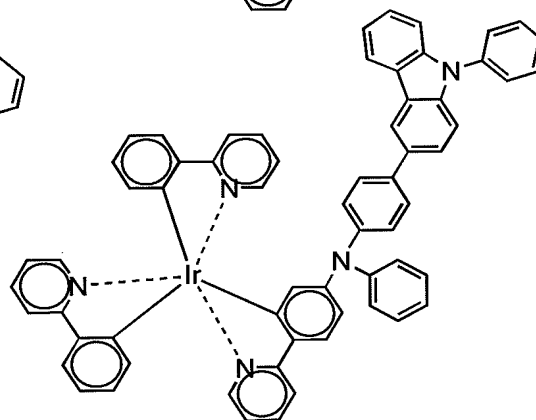
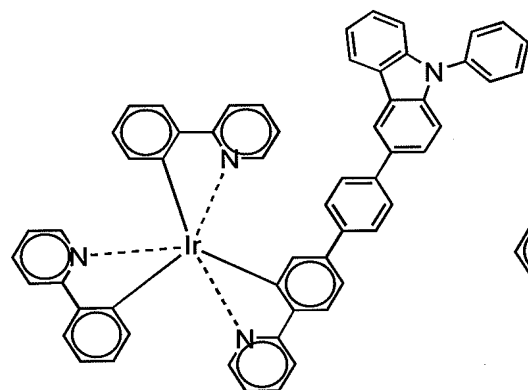
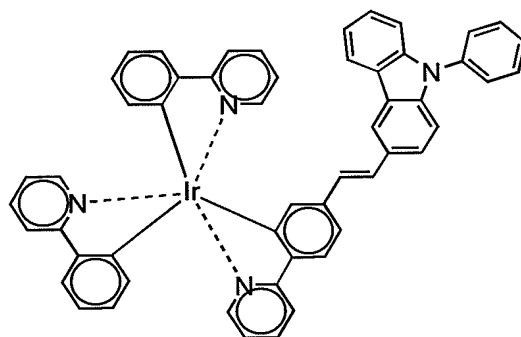
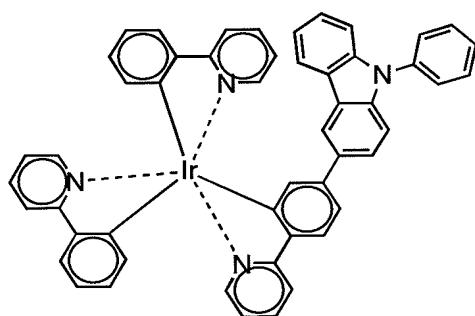
[0064]

Specific examples of the complexes of the present invention are shown below.

[0065]

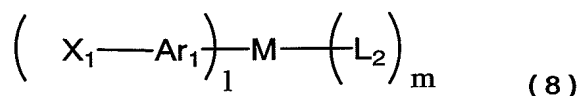


[0066]



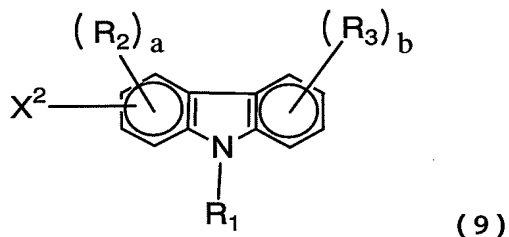
[0067]

The complexes represented by the above formula (1) can be manufactured, for example, by a condensation reaction of a complex represented by the below formula (8) with a carbazole derivative represented by the below formula (9) or (10).



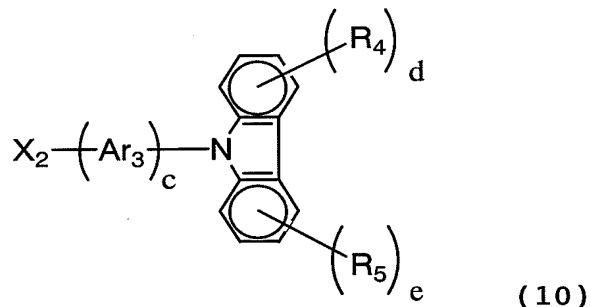
(Here, M, Ar<sub>1</sub>, L<sub>2</sub>, l, and m are the same as those of the above. X<sub>1</sub> represents a halogen atom, sulfoneoxy group, boric acid group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, monohalogenated methyl group, formyl group, cyano group, or vinyl group.)

[0069]



(Here, R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, a, and b are the same as those of the above. X<sub>2</sub> represents a halogen atom, sulfone oxy group, boric acid group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, monohalogenated methyl group, formyl group, cyano group, or vinyl group.)

[0070]



(Here, Ar<sub>3</sub>, R<sub>4</sub>, R<sub>5</sub>, c, d, e, and x<sub>2</sub> are the same as those of the above. )

[0071]

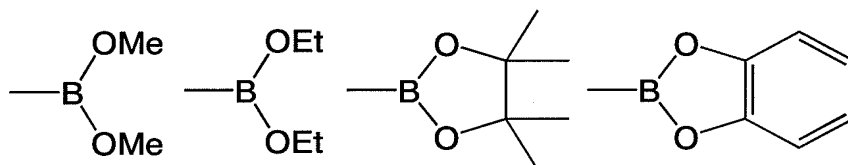
As the halogen atom in  $X_1$  and  $X_2$ , chlorine, bromine, and iodine are exemplified.

[0072]

As the sulfone oxy group, benzene sulfone oxy group, p-toluene sulfone oxy group, methane sulfone oxy group, ethane sulfone oxy group, and trifluoromethane sulfone oxy group are exemplified.

[0073]

As the boric ester group, groups represented by the below formulae are exemplified.



[0074]

As the sulfonium methyl group, groups represented by the below formulae are exemplified.

$-\text{CH}_2\text{SMe}_2\text{X}$  ,  $-\text{CH}_2\text{SPh}_2\text{X}$  (X represents a halogen atom.)

[0075]

As the phosphonium methyl group, a group represented by the below formula is exemplified.

$-\text{CH}_2\text{PPh}_3\text{X}$  (X represents a halogen atom.)

[0076]

As the phosphonate methyl group, a group represented by the below formula is exemplified

$-\text{CH}_2\text{P}(=\text{O})(\text{OR}')_2$

( $\text{R}'$  represents an alkyl group, aryl group, or arylalkyl group.)

[0077]

As the monohalogenated methyl group, chloromethyl group, bromomethyl group, and iodomethyl group are exemplified.

[0078]

Examples of the condensation through a vinylene group include reactions such as: Wittig reaction of a compound having formyl group with a compound having phosphonium methyl group, or a compound having formyl group and phosphonium methyl group;

Heck reaction of a compound having vinyl group with a compound having halogen atom; Knoevenagel reaction of a compound having formyl group with a compound having cyano group; and McMurry reaction of a compound having formyl group, etc.

[0079]

Examples of the formation of a single bond include Suzuki coupling, and Grignard coupling with using nickel catalyst.

[0080]

Concretely, the reactions can be carried out by solving in a organic solvent according to the necessity, with using alkali or an appropriate catalyst, at a temperature of from the melting point to the boiling point.

Known methods can be used described in: Organic Reactions, vol.14, page 270-490, John Wiley & Sons, Inc. (1965); Organic Syntheses, Collective Volume VI, page 407-411, John Wiley & Sons, Inc. (1988); Chem. Rev., vol. 95, page 2457 (1995); J. Organomet. Chem., vol. 576, page 147 (1999); J. Prakt. Chem., vol. 336, page 247 (1994); Makromol. Chem., Macromol.Symp., vol. 12, page 229 (1987), etc.

[0081]

It is preferable that the organic solvent used is subjected to a deoxygenation treatment sufficiently and the reaction is progressed under an inert atmosphere, generally for suppressing a side reaction, though the treatment differs depending on the compound used and the reaction. Further, it is preferable to conduct a dehydration treatment likewise. However, this is not applicable in the case of a reaction in a two-phase system with water, such as a Suzuki coupling reaction.

[0082]

For the reaction, an alkali or suitable catalyst is added appropriately. These may be selected according to the reaction used. It is preferable that the alkali or catalyst is soluble sufficiently in a solvent used for the reaction. As the method of mixing an alkali or catalyst, there is exemplified a method of adding a solution of an alkali or



catalyst slowly while stirring under an inner atmosphere of argon and nitrogen and the like or a method of slowly adding the reaction solution to a solution of an alkali or catalyst, inversely.

[0083]

Although the reaction temperature is not limited, it is usually about -100 to 350 °C, and preferably from 0°C to the boiling point of solvent. Although the reaction time is not limited, it is usually about 30 minutes to 30 hours.

[0084]

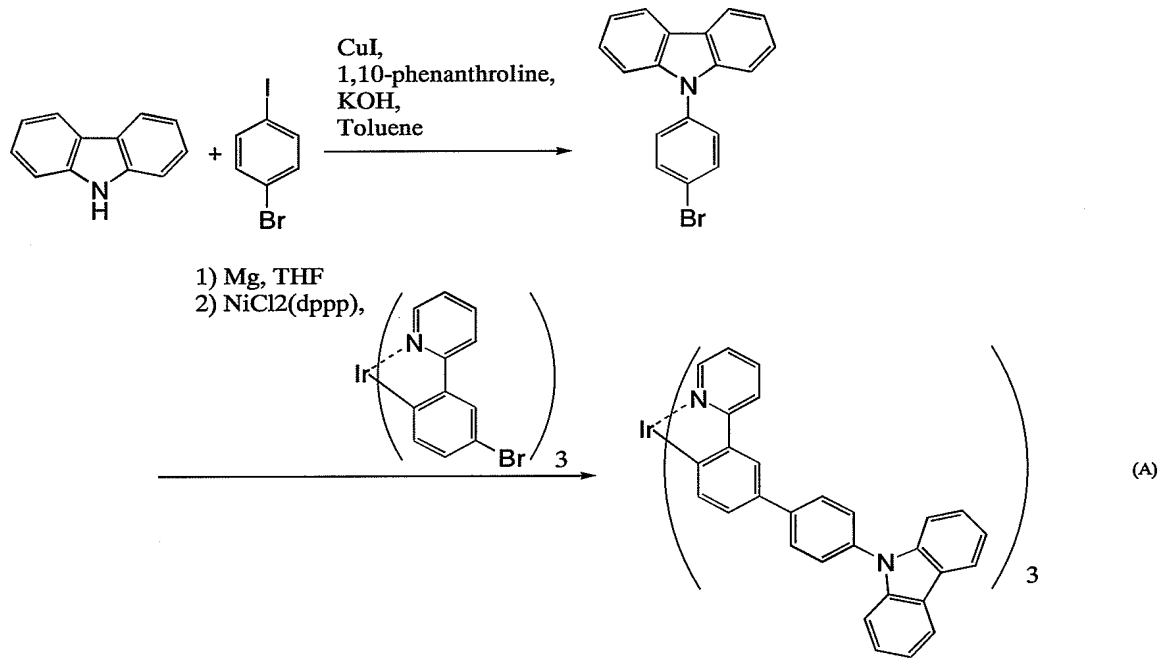
About the extraction and purification of the desired product from a solution of reaction mixture, it differs depending on the complexes, but techniques of usual complex purification, such as recrystallization, sublimation, and chromatography, are used.

[0085]

For example, 1N HCl aqueous solution which is a poor solvent to a complex, is added to deposit the complex, and filtrated to obtain a solid, which is dissolved into an organic solvent, such as dichloromethane or chloroform. This solution is filtrated to remove insoluble materials, concentrated again, and purified by silica gel column chromatography (dichloromethane elution). The fraction solutions of the desired product are collected, and for example, methanol (poor solvent) is added in an appropriate amount, and concentrated to deposit the desired complex, which is filtrated, and dried, and the complex is obtained. The method of producing complex (3) of the present invention is not limited to the above method.

[0086]

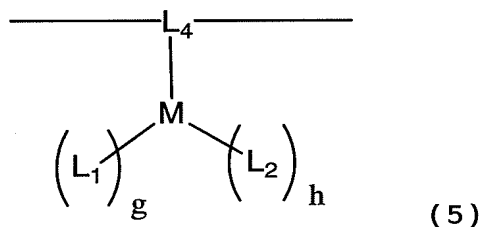
For example, the complex of the present invention represented by the below formula (A) can be produced by the following synthetic route.



[0087]

The polymer compound of the present invention has a repeating unit shown by the below formula (5), (6), or (7), and is characterized by having phosphorescence at a visible region. Two or more kinds of these repeating units may be contained.

[0088]



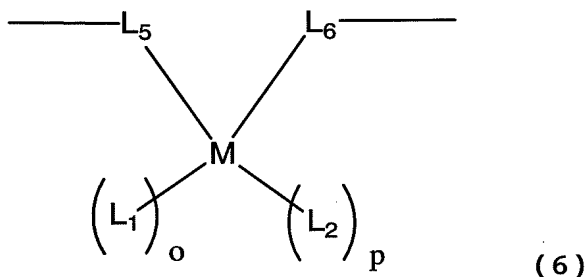
In the formula, M, L<sub>1</sub>, and L<sub>2</sub> are the same as those of the above formula (1). L<sub>4</sub> is a ligand which bonds to M by one or more of nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to two neighboring repeating units with two connecting bonds. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub>s may be mutually same or different. g+h is an

integer of 1-4.)

[0089]

The divalent group represented by  $L_4$  has the same skelton as the ligand represented by  $L_2$ , and has two connecting bonds with the main chain of the polymer instead of substituents.

[0090]

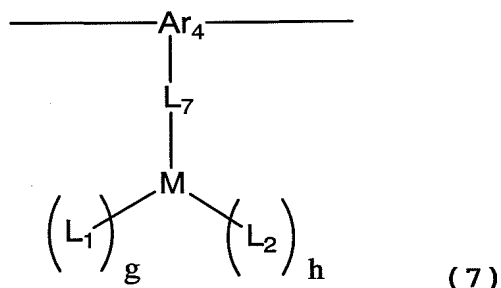


In the formula,  $M$ ,  $L_1$ , and  $L_2$  are respectively the same as those of the above formula (1).  $L_5$  and  $L_6$  are each independently, a ligand which bonds to  $M$  with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has covalent bonds to neighboring repeating unit with two connecting bonds, respectively.  $o$  represents an integer of 1-3 and  $p$  represents an integer of 0-3. When  $p$  is two or more,  $L_2$ s may be the same or different.  $o+p$  is an integer of 1-3.)

[0091]

The monovalent group represented by  $L_5$  and  $L_6$ , has the same skelton as the ligand represented by  $L_2$ , and has one connecting bond with the main chain of the polymer instead of substituents.

[0092]



(wherein,  $M$ ,  $L_1$ , and  $L_2$  are respectively the same as those of the above formula (1).  $Ar_4$  is an arylene group or a divalent heterocyclic group.  $L_7$  is a ligand which bonds to  $M$  with one

or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has a covalent bond to  $Ar_4$  with one connecting bond.  $g$  represents an integer of 1-3 and  $h$  represents an integer of 0-3. When  $h$  is two or more,  $L_2s$  may be the same or different.  $g+h$  is an integer of 1-4.).

[0093]

The group represented by  $Ar_4$ , is a main-chain skelton of the polymer, and is a group having a covalent bond to  $L_7$ .

[0094]

The monovalent group represented by  $L_7$ , has the same skelton as the ligand represented by  $L_2$ , and has one connecting bond to  $Ar_4$  instead of a substituent.

[0095]

The repeating unit shown by the above formula (5) or (7) is usually

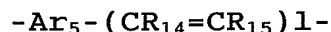
0.01-50 % by mole based on the total mole number of all the repeating units in which the polymer compound of the present invention has, and preferably, it is 0.1-10 % by mole.

[0096]

The polymer compound of the present invention can contain a repeating unit other than the repeating unit shown by the above formula (5) or (7). As the repeating unit, the below formula (11) or (12) is exemplified. It is preferable that the repeating unit represented by formula (12) is contained in view of the device life time.

[0097]

Formula (11)



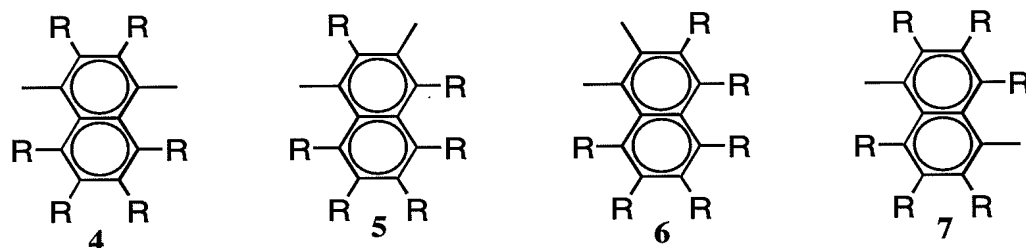
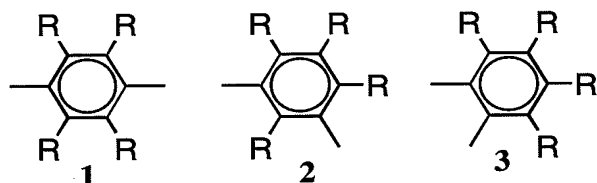
In the formula,  $Ar_5$  is an arylene group or a divalent heterocyclic group.

[0098]

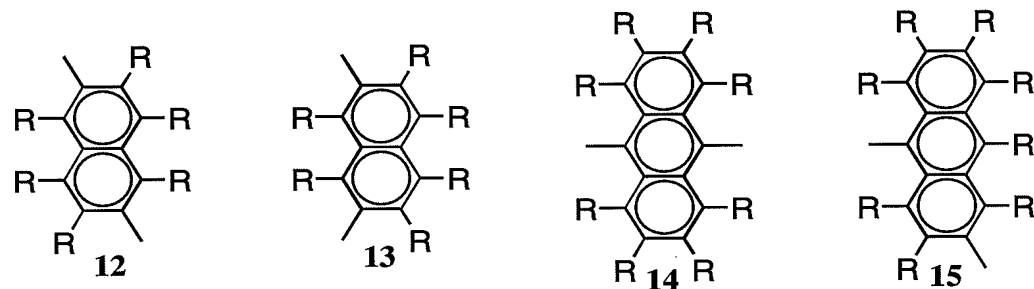
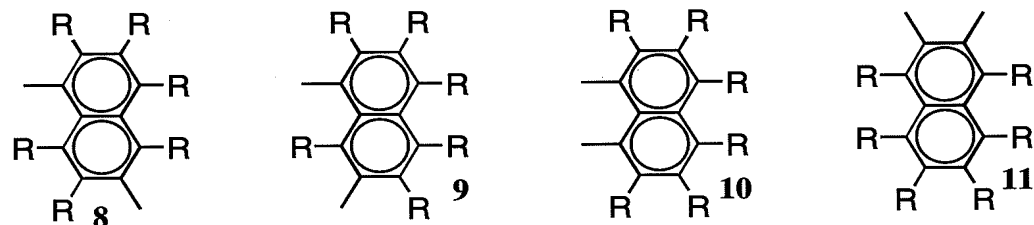
Here, the arylene group has usually 6-60, preferably 6-20 carbon atoms, and examples thereof include phenylene group (for example, following formulas 1-3), naphthalenediyl group (following formulas 4-13), anthracenylene group (following formulas 14-19), biphenylene group (following formulas 20-25),

triphenylene group (following formulas 26-28), condensed-ring compound group (following formulas 29-38), etc. Here, the number of carbon atoms of substituent R is not counted as the number of carbon atoms of arylene group.

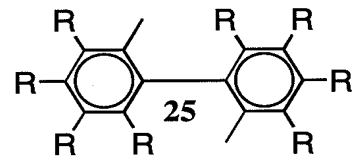
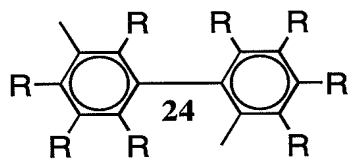
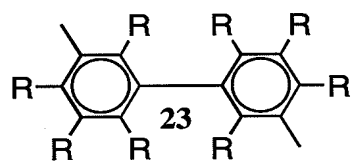
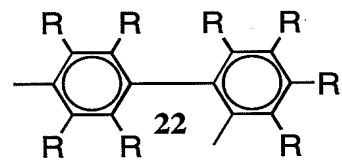
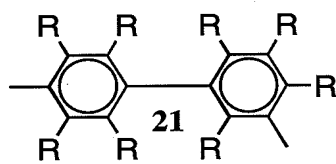
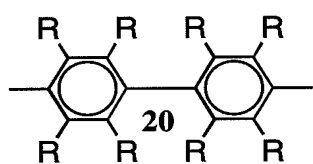
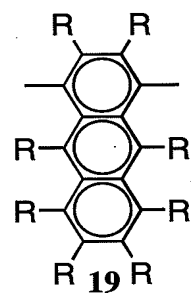
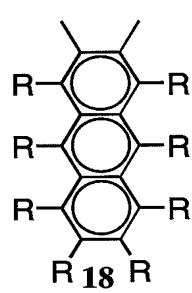
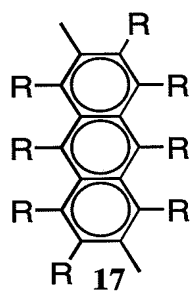
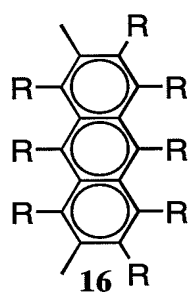
[0099]



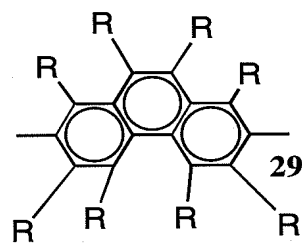
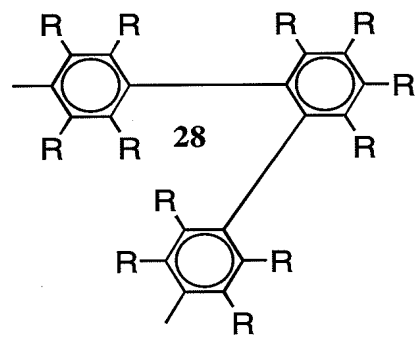
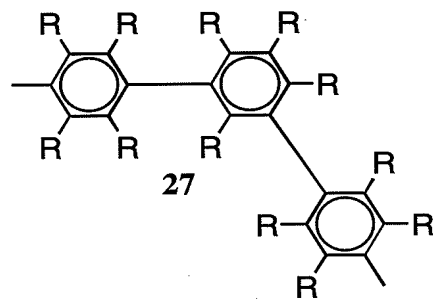
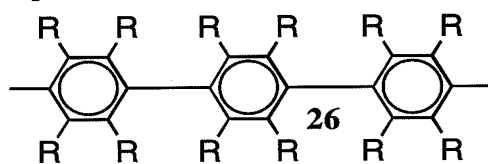
[0100]



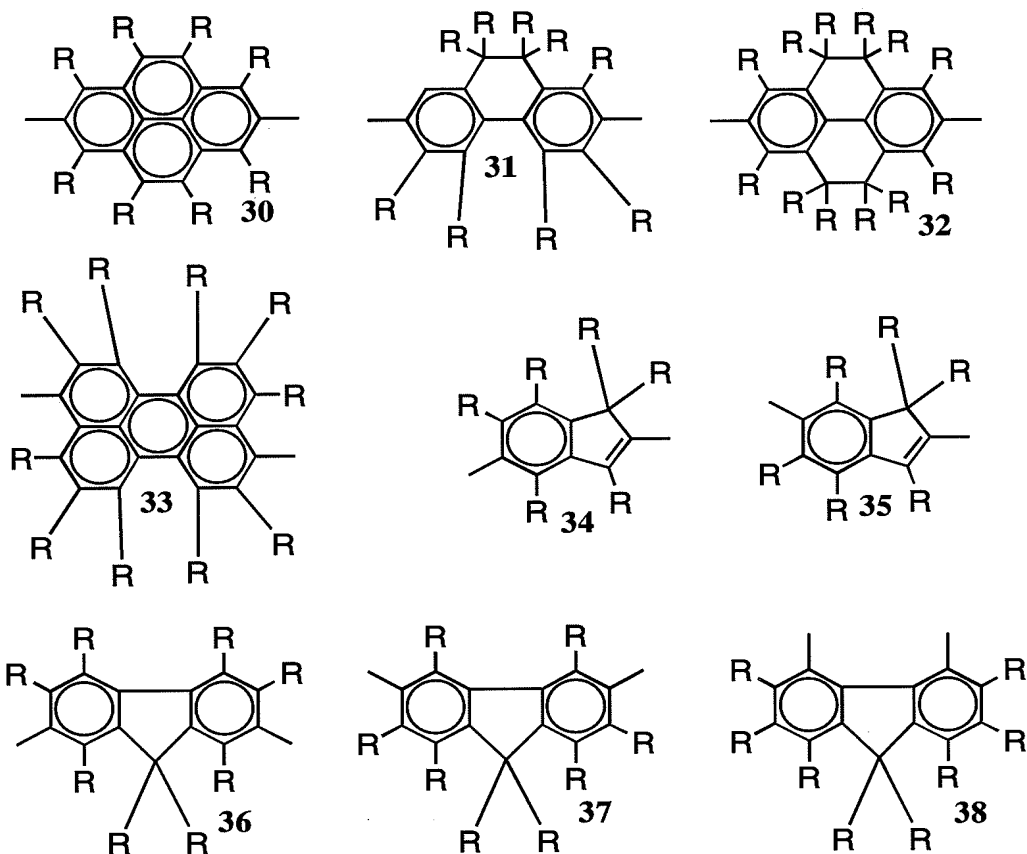
[0101]



[0102]



[0103]



[0104]

In the present invention, the divalent heterocyclic group means an atomic group in which two hydrogen atoms are removed from a heterocyclic compound, and the number of carbon atoms is usually 4-60, and preferably 4-20. Here, the number of carbon atoms of substituent is not counted as the number of carbon atoms of the divalent heterocyclic group.

[0105]

The heterocyclic compound means an organic compound having a cyclic structure in which at least one heteroatom such as oxygen, sulfur, nitrogen, phosphorus, boron, etc. is contained in the cyclic structure as the element other than carbon atoms.

[0106]

Examples of the divalent heterocyclic group include followings.

[0107]

Divalent heterocyclic groups containing nitrogen as a hetero atom; pyridine-diyl group (following formulas 39-44), diaza phenylene group (following formulas 45-48), quinolinediyl group (following formulas 49-63), quinoxalinediyl group (following formulas 64-68), acridinediyl group (following formulas 69-72), bipyridyldiyl group (following formulas 73-75), phenanthrolinediyl group (following formulas 76-78), etc.

Groups having a fluorene structure containing silicon, nitrogen, sulfur, selenium, etc. as a hetero atom (following formulas 79-93). It is preferable to have an aromatic amine monomer containing a nitrogen atom, such as carbazole of formulas 82-84 or triphenylaminediyl group, in view of light emitting efficiency.

[0108]

5 membered heterocyclic groups containing silicon, nitrogen, sulfur, selenium, etc. as a hetero atom: (following formulas 94-98).

[0109]

Condensed 5 membered heterocyclic groups containing silicon, nitrogen, sulfur, selenium, etc. as a hetero atom: (following formulas 99-109), benzothiadiazole-4,7-diyl group, benzo oxadiazole-4,7-diyl group, etc.

[0110]

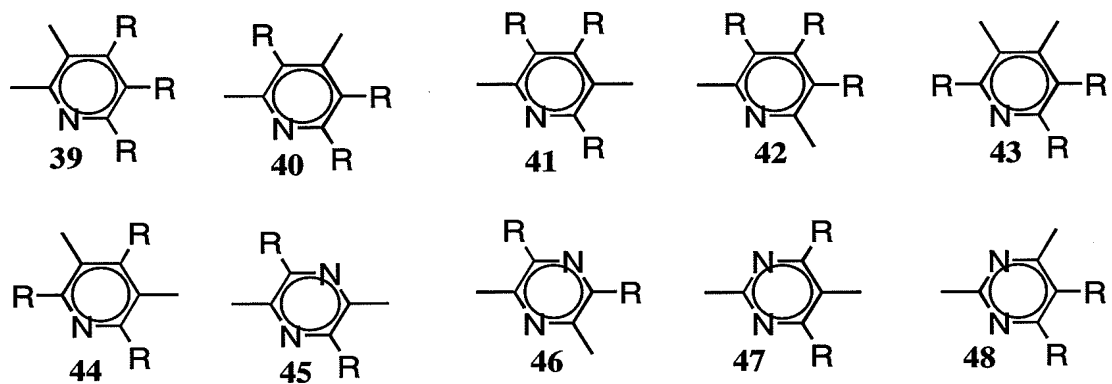
5 membered heterocyclic groups containing silicon, nitrogen, sulfur, selenium, etc. as a hetero atom, which are connected at the a position of the hetero atom to form a dimer or an oligomer (following formulas 110-118); and

[0111]

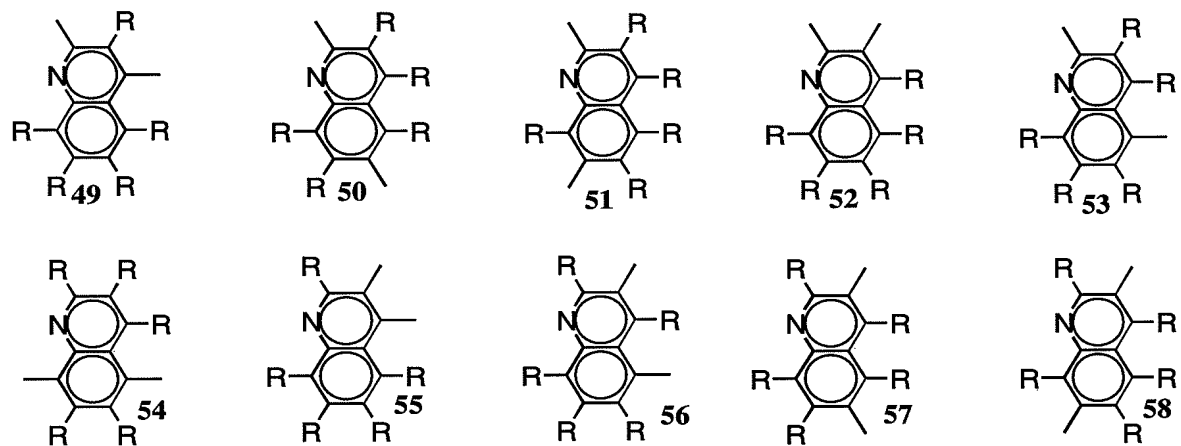
5 membered ring heterocyclic groups containing silicon, nitrogen, oxygen, sulfur, selenium, as a hetero atom is connected with a phenyl group at the a position of the hetero atom (following formulas 112-118).

[0112]

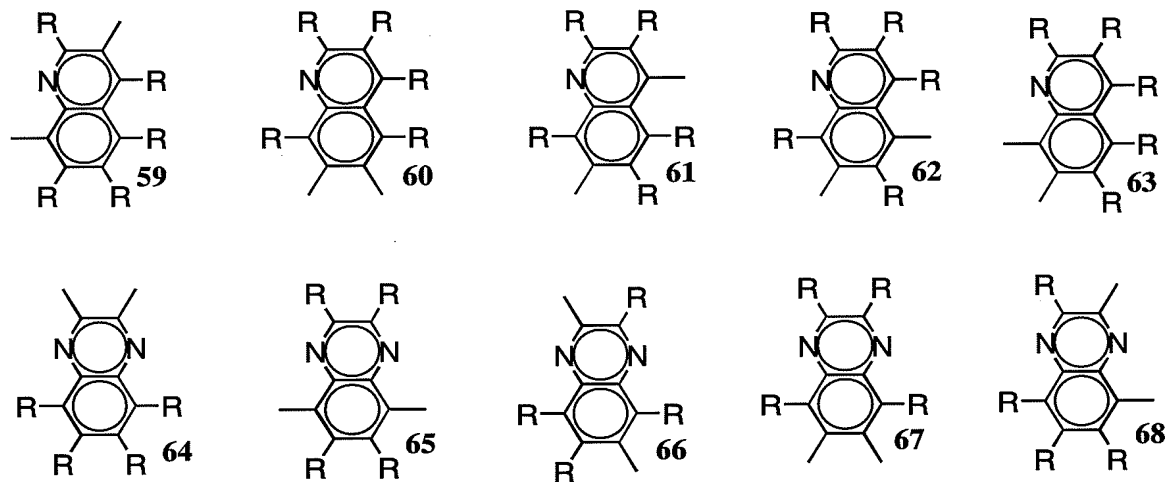




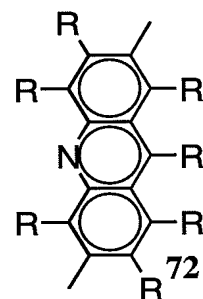
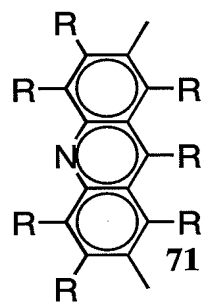
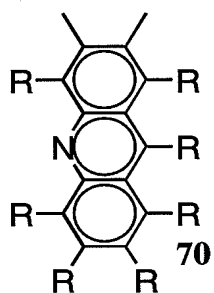
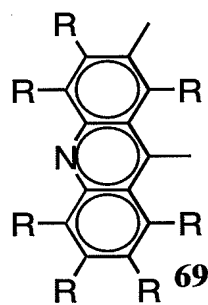
[0113]



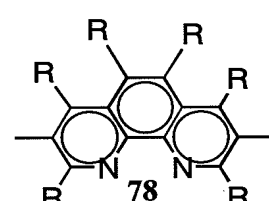
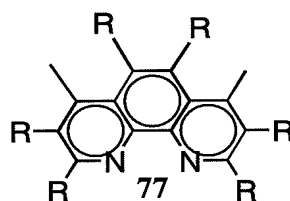
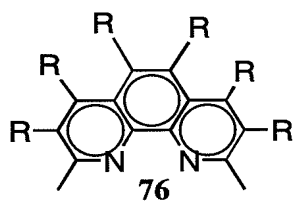
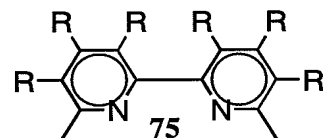
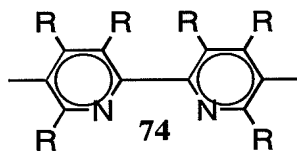
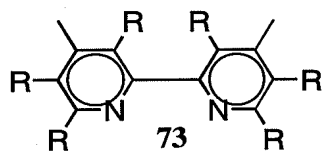
[0114]



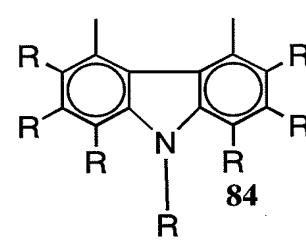
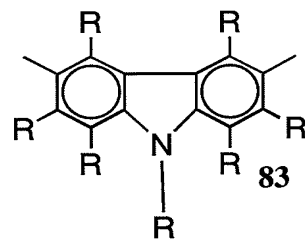
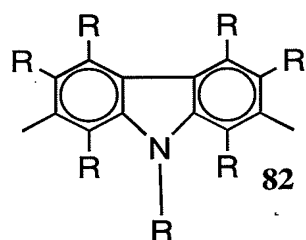
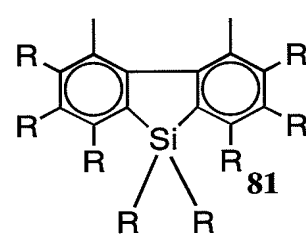
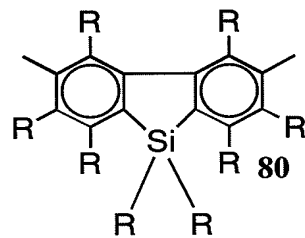
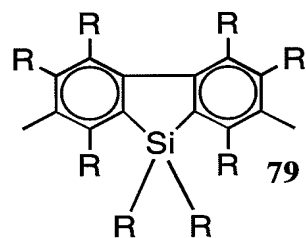
[0115]



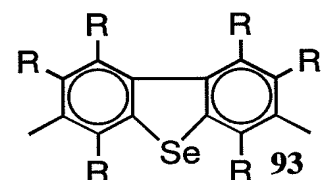
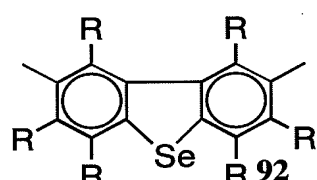
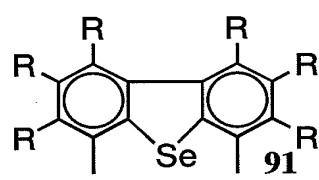
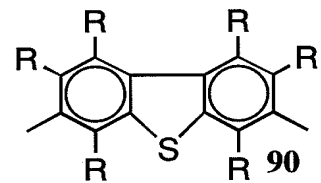
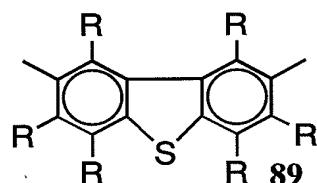
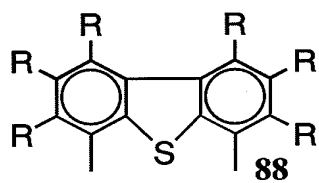
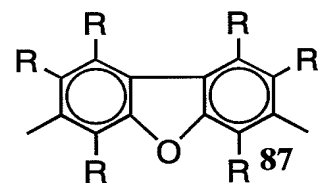
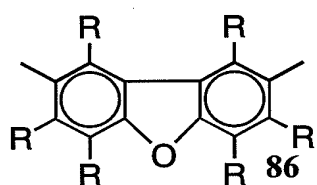
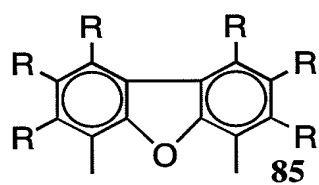
[0116]



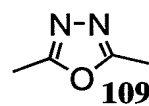
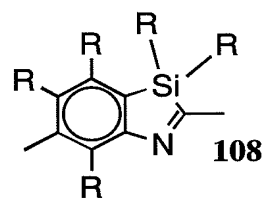
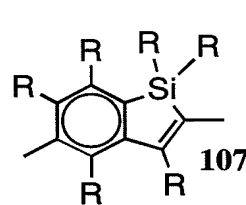
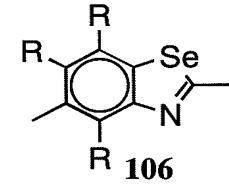
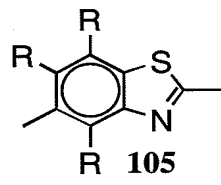
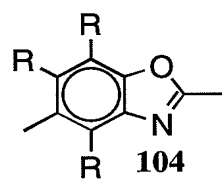
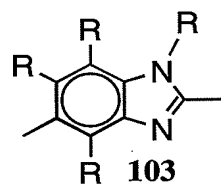
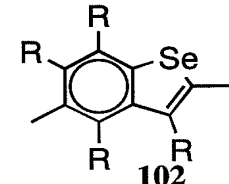
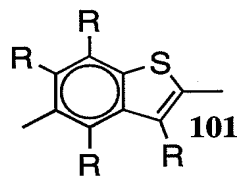
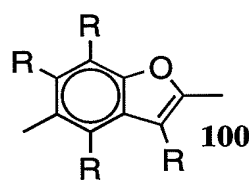
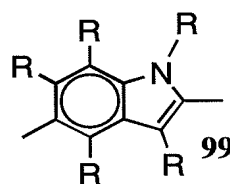
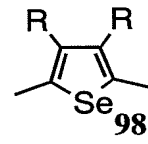
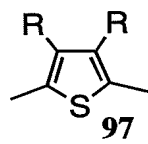
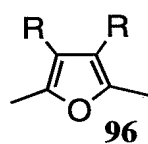
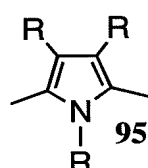
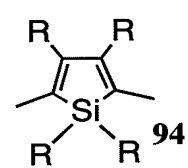
[0117]



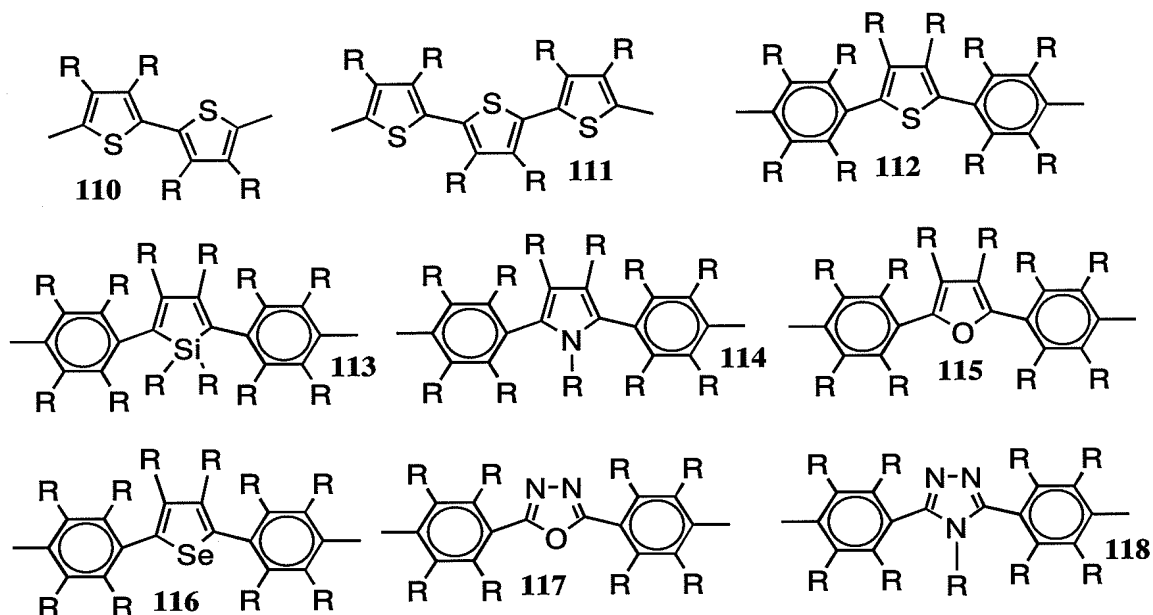
[0118]



[0119]



[0120]



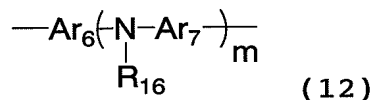
[0121]

In the formula, R represents each independently a hydrogen atom, halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkylsilyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, aryl alkoxy group, aryl alkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyl oxy group, imino group, amide group, aryl alkenyl group, aryl alkynyl group, cyano group, or monovalent heterocyclic group. Rs may be mutually connected to form a ring. In order to improve the solubility in a solvent, it is preferable that at least one of the R contains an alkyl group of long chain. Specifically, the groups of the above R<sub>2</sub> and R<sub>3</sub> are exemplified.

[0122]

R<sub>14</sub> and R<sub>15</sub> each independently represent a hydrogen atom, alkyl group, aryl group, monovalent heterocyclic group, or cyano group. l is 0 or 1. As the alkyl group, aryl group, and a monovalent heterocyclic group, it exemplifies In the group represented by R<sub>14</sub> or R<sub>15</sub>, the groups described in the above R<sub>2</sub> and R<sub>3</sub> are exemplified.

[0123]

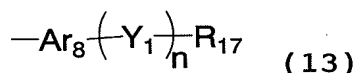


In the formula, Ar<sub>6</sub> and Ar<sub>7</sub> each independently represent an arylene group or a divalent heterocyclic group. R<sub>16</sub> represents an alkyl group, aryl group, arylalkyl group, arylalkenyl group, aryl alkynyl group or a monovalent heterocyclic group, the group represented by the following (13), or the group represented by the following (14). m is an integer of 1-4.

[0124]

In the group represented by R<sub>16</sub>, as the alkyl group, aryl group, arylalkyl group, aryl alkenyl group, aryl alkynyl group, and a monovalent heterocyclic group, the group described in the above R<sub>2</sub> and R<sub>3</sub> are exemplified.

[0125]

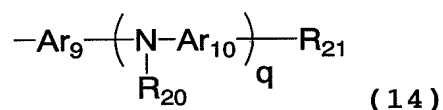


In the formula, Ar<sub>8</sub> is an arylene group or a divalent heterocyclic group. Specifically, the groups described in the above Ar<sub>3</sub> are exemplified

[0126]

R<sub>17</sub> represents a hydrogen atom, alkyl group, aryl group, a monovalent heterocyclic group, or the group shown by the below formula (14). Y<sub>1</sub> represents —CR<sub>18</sub>=CR<sub>19</sub>—, or —C=C—. R<sub>18</sub> and R<sub>19</sub> each independently represent a hydrogen atom, alkyl group, aryl group, a monovalent heterocyclic group, or cyano group. n is an integer of 0-2. In the group shown by R<sub>17</sub>, R<sub>18</sub> or R<sub>19</sub>, as the alkyl group, aryl group, and monovalent heterocyclic group, the groups described in the above R<sub>2</sub> and R<sub>3</sub> are exemplified.

[0127]



In the formula, Ar<sub>9</sub> and Ar<sub>10</sub> each independently represent an arylene group or a divalent heterocyclic group. R<sub>20</sub> represents an alkyl group, aryl group, arylalkyl group, arylalkenyl group, arylalkynyl group, or a monovalent heterocyclic group. R<sub>21</sub> represents a hydrogen atom, alkyl group, aryl group, or a monovalent heterocyclic group. q is an integer of 1-4.

[0128]

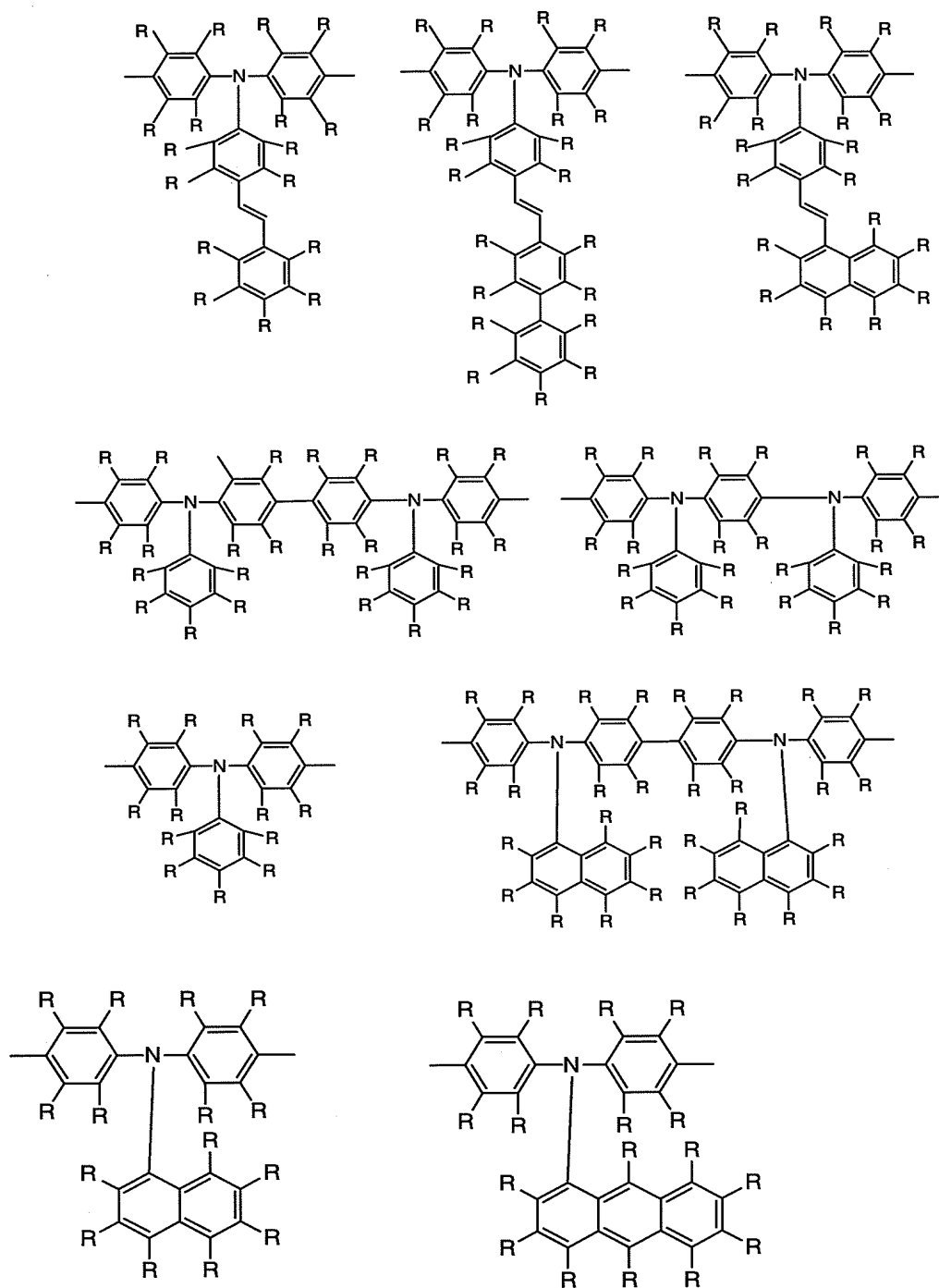
In the group represented by Ar<sub>9</sub> and Ar<sub>10</sub>, as the arylene group and divalent heterocyclic group, the groups described in the above Ar<sub>3</sub> are exemplified.

[0129]

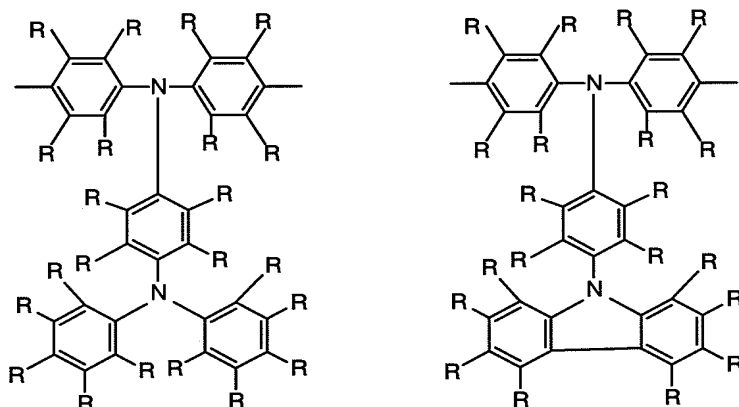
In the group represented by R<sub>20</sub> and R<sub>21</sub>, as the alkyl group, aryl group, arylalkyl group, aryl alkenyl group, aryl alkynyl group, and a monovalent heterocyclic group, the groups described in the above R<sub>2</sub> and R<sub>3</sub> are exemplified.

[0130]

As the preferable example of the repeating unit represented by the above formula (12), those of the following are exemplified.



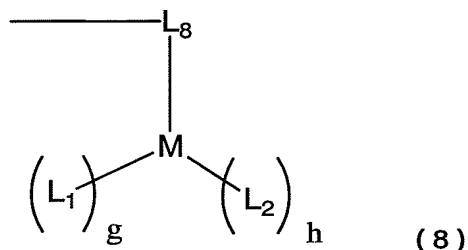
[0131]



Here, R each independently represents a hydrogen atom, halogen atom, alkyl group, alkoxy group, alkylthio group, alkyl amino group, alkyl silyl group, aryl group, aryloxy group, arylthio group, aryl amino group, aryl silyl group, arylalkyl group, aryl alkoxy group, aryl alkylthio group, arylalkyl amino group, arylalkyl silyl group, acyl group, acyl oxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or monovalent heterocyclic group. Rs may be connected mutually to form a ring. In order to improve the solubility in a solvent, it is preferable that at least one R contains an alkyl group of long chain. Specifically, the groups described in R<sub>2</sub> and R<sub>3</sub> are exemplified.

[0132]

The polymer compound of the present invention has a structure represented by the below formula (8) and has phosphorescence in a visible region. In this case, it is preferable in view of the light emitting efficiency.



Here, M, L<sub>1</sub>, L<sub>2</sub> are respectively the same as those of the above formula (1). L<sub>8</sub> is a ligand which bonds to M with one or more nitrogen atom, oxygen atom, carbon atom, sulfur atom, or phosphorus atom, and has a covalent bond at the polymer terminal



with one connecting bond. g represents an integer of 1-3 and h represents an integer of 0-3. When h is two or more, L<sub>2</sub> may be the same or different. g+h is an integer of 1-4.)

[0133]

As the ligand represented by L<sub>8</sub>, it is a ligands represented by the above L<sub>2</sub>, and has a covalent bond to the terminal of a polymer main-chain at an arbitrary position.

[0134]

The polymer of the present invention may also be a random, block or graft copolymer, or a polymer having an intermediate structure thereof, for example, a random copolymer having block property. From the viewpoint for obtaining a polymeric fluorescent substance having high fluorescent quantum yield, random copolymers having block property and block or graft copolymers are more preferable than complete random copolymers. Further, a polymer having a branched main chain and more than three terminals, and a dendrimer may also be included.

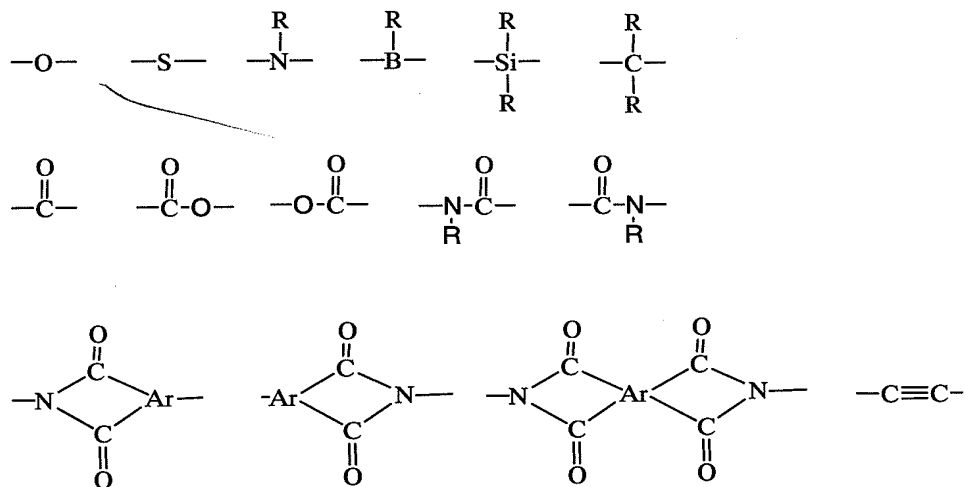
[0135]

The end group of polymeric compound may also be protected with a stable group since if a polymerization active group remains intact, there is a possibility of reduction in light emitting property and life-time when made into an device. Those having a conjugated bond continuing to a conjugated structure of the main chain are preferable, and there are exemplified structures connected to an aryl group or heterocyclic compound group via a carbon-carbon bond. Specifically, the substituents described as Chemical Formula 10 in JP-A-9-45478, etc. are exemplified.

[0136]

The polymer compound of the present invention may be connected by non-conjugated units within a range of not injuring the luminescence characteristic or the charge transportating characteristic, and the non-conjugated portions thereof may be contained in the repeating unit. Examples thereof include: groups shown below, combinations of groups shown below with vinylene groups; and combinations two or more of the following groups. Here, R is the same as

that of the above, and Ar has 6-60 carbon atoms, and represents an aromatic compound group having a suitable connecting bond.  
[0137]



[0138]

The polymer compound of the present invention has usually polystyrene reduced number average molecular weights of  $10^3$ - $10^8$ .

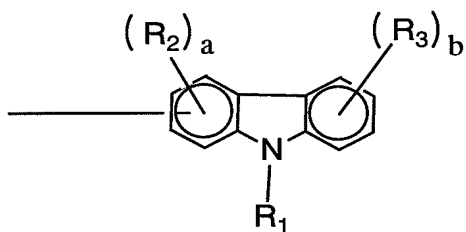
The polymer compound of the present invention is preferably a conjugated type polymer compound. Here, the conjugated type polymer compound means a polymer compound where delocalized  $\pi$  electron pair exist along with the main-chain of the polymer compound. As the delocalized electron, a unpaired electron or a lone electron pair may join to the resonance instead of a double bond.

The polymer compound of the present invention has the advantage that a layer can be formed by an application method. On the other hand, the complex of the present invention can form a layer preferably by a depositing method.

[0139]

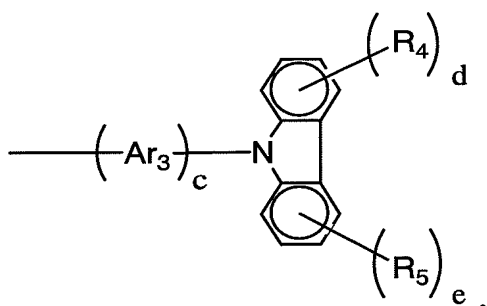
The complex and the polymer compound of the present invention usually have a structure derived from triplet light-emitting complex. That is, the complex of the present invention is a triplet light-emitting complex, and the following substituent is contained in the ligand.

(B)



or

(C)



Furthermore, the polymer compound of the present invention is a polymer compound having a structure derived from a complex which is a triplet light-emitting complex, and contains a ligand having the substituent of the above (B) or (C).

[0140]

The triplet light-emitting complexes are those having been used as a low molecular weight EL material. Such materials are disclosed, for example, in: Nature, (1998) 395,151; Appl. Phys. Lett., (1999) 75(1), 4; Proc. SPIE-Int. Soc. Opt. Eng., (2001) 4105 ; (Organic Light-Emitting Materials and Devices IV), 119; J. Am. Chem. Soc., (2001) 123, 4304; Appl. Phys. Lett., (1997) 71(18), 2596; Syn. Met., (1998) 94(1), 103; Syn. Met., (1999) 99(2), 1361; and Adv. Mater., (1999), 11 (10), 852.

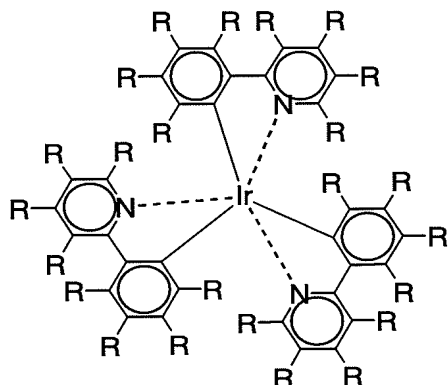
[0141]

In the below examples, those where at least one R in each of the complexes is a substituent represented by the above (B) or (C) are the complexes of the present invention.

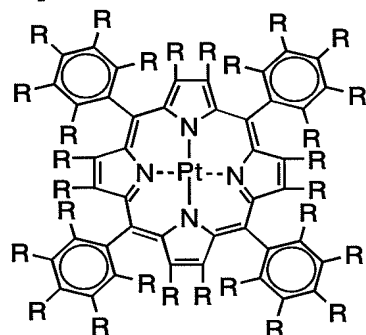
Moreover, a polymer compound in which at least one R in each of the complexes is a substituent represented by the above (B) or (C), and at least one R bonds to the polymer as a

connecting group, are the polymer compounds of the present invention.

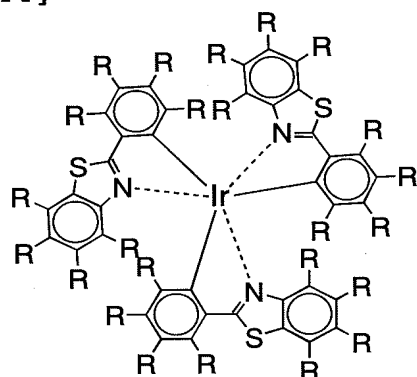
[0142]



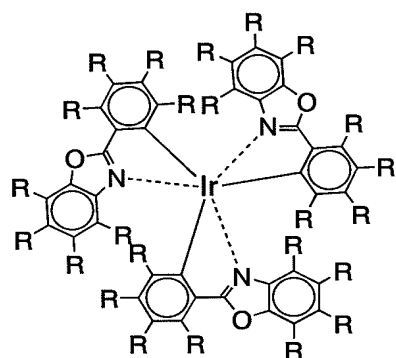
[0143]



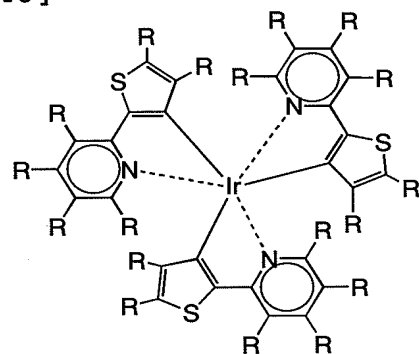
[0144]



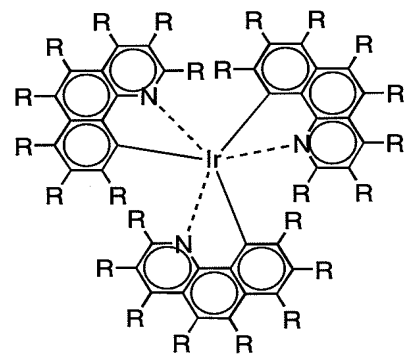
[0145]



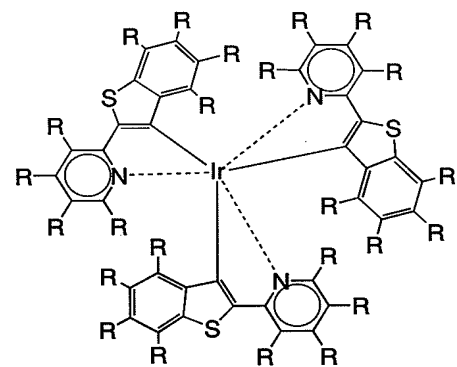
[0146]



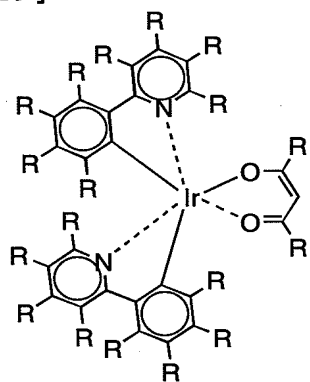
[0147]



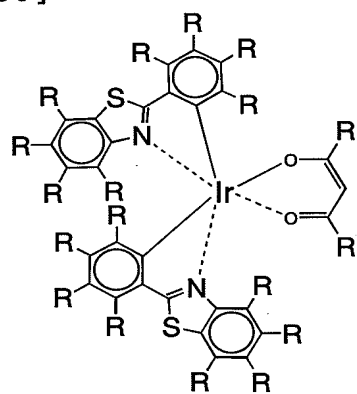
[0148]



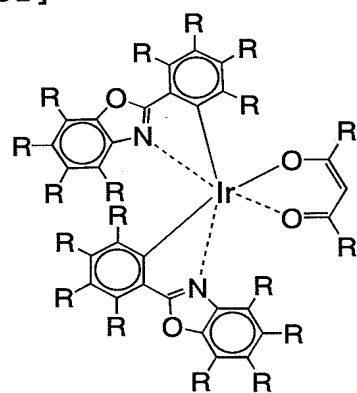
[0149]



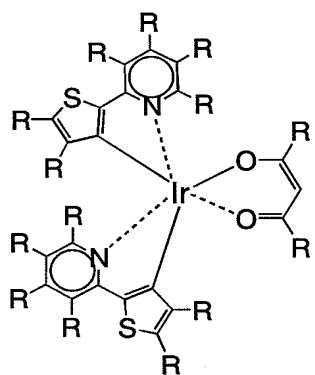
[0150]



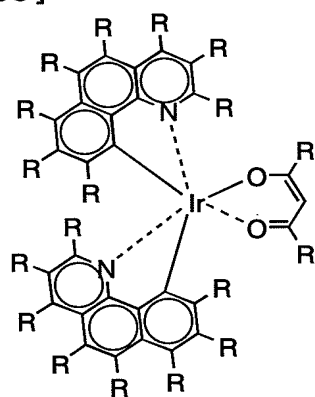
[0151]



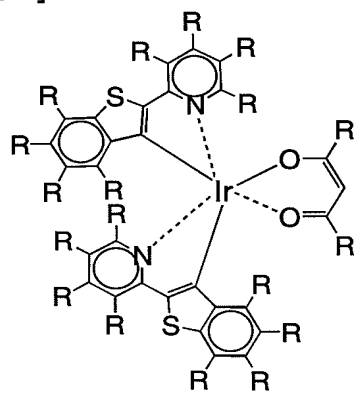
[0152]



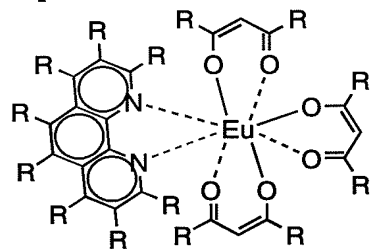
[0153]



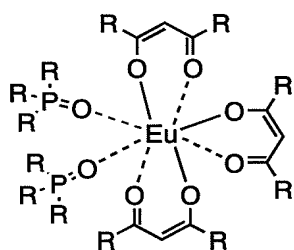
[0154]



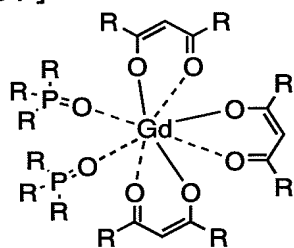
[0155]



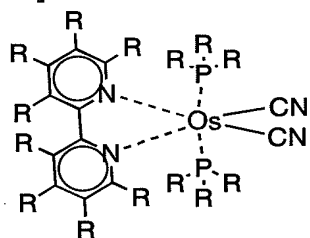
[0156]



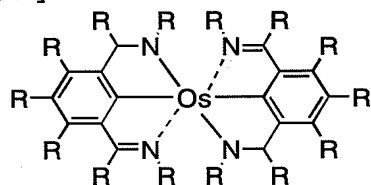
[0157]



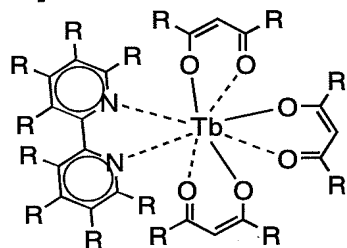
[0158]



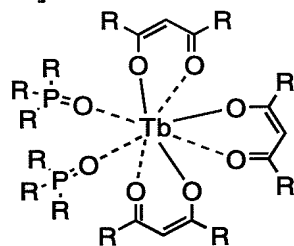
[0159]



[0160]

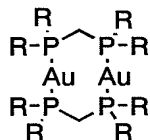


[0161]





[0162]



[0163]

Here, R each independently represent a hydrogen atom, halogen atom, alkyl group, alkoxy group, alkylthio group, alkylamino group, alkyl silyl group, aryl group, aryloxy group, arylthio group, arylamino group, arylsilyl group, arylalkyl group, arylalkoxy group, arylalkylthio group, arylalkylamino group, arylalkylsilyl group, acyl group, acyloxy group, imino group, amide group, arylalkenyl group, arylalkynyl group, cyano group, or monovalent heterocyclic group. Rs may be mutually connected to form a ring. In order to improve the solubility into a solvent, it is preferable that at least one R contain a long chain alkyl group.

[0164]

Next, the manufacture method of the polymer compound of the present invention is explained.

When the polymer compound of the present invention has vinylene group in the main chain, the method described in JP-A-5-202355 is exemplified. Namely, exemplified are: a polymerization by Wittig reaction of a compound having formyl group with a compound having phosphonium-methyl group, or of compounds which have a formyl group and a phosphonium-methyl group; a polymerization by Heck reaction of a compound having a vinyl group with a compound having a halogen atom; a polycondensation by a dehydrohalogenation method of a compound having two or more monohalogenated-methyl group; a polycondensation by a sulfonium-salt decomposition method of a compound having two or more sulfonium-methyl groups; a polymerization by Knoevenagel reaction of a compound having a formyl group with a compound having a cyano group; a polymerization by McMurry reaction of a compound having two or more formyl groups, etc.

When the polymer compound of the present invention has triple bond in the main chain, Heck reaction can be used, for example.

[0165]

When there contained neither vinylene group nor triple bond in the main chain, exemplified are: a polymerization method by Suzuki coupling reaction from the corresponding monomers; a polymerization method by Grignard reaction; a polymerization method by Ni(0) catalyst; a polymerization method by an oxidizer, such as FeCl<sub>3</sub>; an electrochemically oxidization polymerization; and a method by decomposition of an intermediate polymer having an suitable elimination group.

[0166]

Among these, a polymerization by Wittig reaction, a polymerization by Heck reaction, a polymerization by Knoevenagel reaction, polymerization method by Suzuki coupling reaction, a polymerization by Grignard reaction, a polymerization by Ni(0) catalyst are preferable, since it is easy to carry out structure control.

[0167]

Specifically, a compound having a plurality of reactive substituents is dissolved in an organic solvent, according to necessity, and can be reacted using alkali or appropriate catalyst, at the temperature between the boiling point and the melting point of the organic solvent of the organic solvent.

Known methods which can be used are described in: Organic Reactions, Volume 14, page 270-490, John Wiley & Sons, Inc., 1965 ; Organic Reactions, Volume 27, page 345-390, John Wiley & Sons, Inc., 1982; Organic Syntheses, Collective Volume VI, page 407-411, John Wiley & Sons, Inc., 1988; Chemical Review (Chem.Rev.), Volume 95, page 2457 (1995); Journal of Organometallic Chemistry (J.Organomet.Chem.), Volume 576, page 147 (1999); Journal of Praktical Chemistry (J.Prakt.Chem.), Volume 336, page 247 (1994); and Macromolecular Chemistry, Macromolecular Symposium (Makromol. Chem., Macromol.Symp.), Volume 12th, page 229 (1987).

[0168]

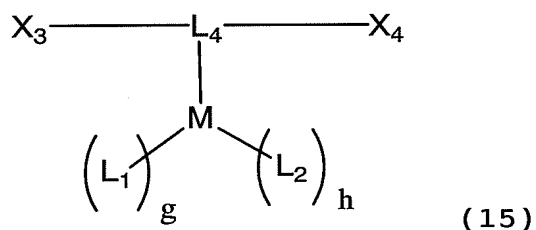
It is preferable that the organic solvent used is subjected to a deoxygenation treatment sufficiently and the reaction is progressed under an inert atmosphere, generally for suppressing a side reaction, though the treatment differs depending on compounds and reactions used. Further, it is preferable to conduct a dehydration treatment likewise. However, this is not applicable in the case of a reaction in a two-phase system with water, such as a Suzuki coupling reaction.

[0169]

For the reaction, alkali and a suitable catalyst is added. They can be selected according to the reaction to be used. It is preferable that the alkali or the catalyst can be dissolved in a solvent used for a reaction. Example of the method for mixing the alkali or the catalyst, include a method of adding a solution of alkali or a catalyst slowly, to the reaction solution with stirring under an inert atmosphere of argon, nitrogen, etc. or conversely, a method of adding the reaction solution to the solution of alkali or a catalyst slowly.

[0170]

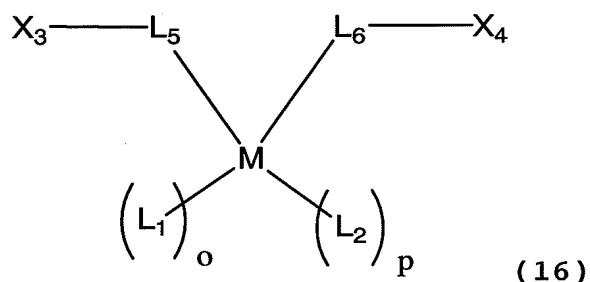
The polymer compound represented by the above formula (5) can be suitably manufactured by carrying out condensation polymerization of the monomer shown by the below formula (15).



(In the formula, M, L<sub>1</sub>, L<sub>2</sub>, L<sub>4</sub>, g, and h are the same as those of formula (5). X<sub>3</sub> and X<sub>4</sub> each independently represent a halogen atom, sulfone oxy group, boric-acid group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, monohalogenated methyl group, formyl group, cyano group, or vinyl group.)

[0171]

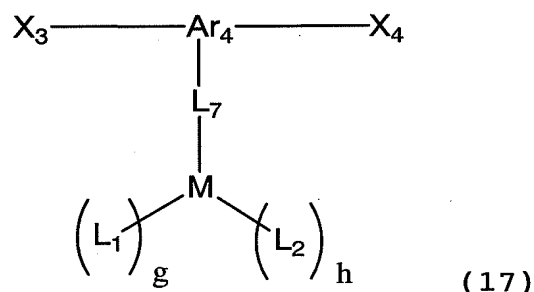
The polymer compound represented by the above formula (6) can be suitably manufactured by carrying out condensation polymerization of the monomer shown by the below formula (16).



(In the formula, M, L<sub>1</sub>, L<sub>2</sub>, L<sub>5</sub>, L<sub>6</sub>, X<sub>3</sub>, X<sub>4</sub>, o, and p are the same as those of the above.)

[0172]

The polymer compound represented by the above formula (7) can be suitably manufactured by carrying out condensation polymerization of the monomer shown by the below formula (17).



(Here, M, L<sub>1</sub>, L<sub>2</sub>, L<sub>7</sub>, Ar<sub>4</sub>, X<sub>3</sub>, X<sub>4</sub>, g, and h are the same as those of the above.)

[0173]

Among the groups shown by X<sub>3</sub> and X<sub>4</sub>, as the halogen atom, sulfone oxy group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, and monohalogenated methyl group, the compounds described in the above X<sub>1</sub> and X<sub>2</sub> are exemplified.

[0174]

When the polymer compound of the present invention has repeating units other than the repeating unit of formula (5) - (7), it can be produced by copolymerizing other monomers as

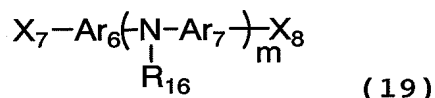
repeating units other than repeating units of formula (5)-(7).  
[0175]

As the monomer used as repeating unit other than the repeating unit of formulas (5) - (7), the compounds of the below formula (18) and (19) are exemplified.



In the formula,  $Ar_5$ ,  $R_{14}$ ,  $R_{15}$ , and 1 are the same as those of the above.  $X_5$  and  $X_6$  each independently represent a halogen atom, sulfone oxy group, boric acid group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, monohalogenated methyl group, formyl group, cyano group, or vinyl group.

[0176]



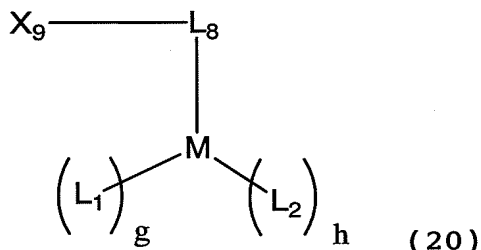
In the formula,  $Ar_6$ ,  $Ar_7$ ,  $R_{16}$ , and m are the same as those of the above.  $X_7$  and  $X_8$  each independently represent a halogen atom, sulfone oxy group, boric acid group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, monohalogenated methyl group, formyl group, cyano group, or vinyl group.

[0177]

Among the groups shown by  $X_5$  to  $X_8$ , as the halogen atom, sulfone oxy group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, and monohalogenated methyl group, the compounds described in the above  $X_1$  and  $X_2$  are exemplified.

[0178]

The polymer compound of the present invention can be manufactured by copolymerizing a monomer shown by the below formula (20) in existence of a monomer of formula (18) or formulas (19).



In the formula, M, L<sub>1</sub>, L<sub>2</sub>, L<sub>8</sub>, g, and h are the same as those of the above. X<sub>9</sub> represents a halogen atom, sulfone oxy group, boric acid group, boric ester group, sulfonium methyl group, phosphonium methyl group, phosphonate methyl group, monohalogenated methyl group, formyl group, cyano group, or vinyl group.

[0179]

When a polymeric light-emitting substance comprising the polymer compound of the present invention is used for a light-emitting materials of a polymer LED, the purity thereof exerts an influence on light emitting property, therefore, it is preferable that a monomer is purified by a method such as distillation, sublimation purification, re-crystallization and the like before being polymerized. Further, it is preferable to conduct a purification treatment such as re-precipitation purification, chromatographic separation and the like after the polymerization.

[0180]

Next, the polymer LED of the present invention will be explained. The polymer LED of the present invention comprises an light emitting layer between the electrodes consisting of an anode and a cathode, and the light emitting layer contains the complex or the polymer compound of the present invention.

[0181]

As the polymer LED of the present invention, exemplified are: a polymer LED having an electron transporting layer between a cathode and a light emitting layer; a polymer LED having an hole transporting layer between an anode and a light emitting layer; and a polymer LED having an electron transporting layer between an cathode and a light emitting layer, and a hole transporting layer between an anode and a

light emitting layer.

Also exemplified are: a polymer LED having a layer containing a conductive polymer between at least one of the electrodes and a light emitting layer adjacently to the electrode; and a polymer LED having a buffer layer having a mean thickness of 2nm or less between at least one of the electrodes and a light emitting layer adjacently to the electrode.

[0182]

For example, the following structures a) to d) are specifically exemplified.

- a) anode/light emitting layer/cathode
- b) anode/hole transporting layer/light emitting layer/cathode
- c) anode/light emitting layer/electron transporting layer/cathode
- d) anode/hole transporting layer/light emitting layer/electron transporting layer/cathode

(wherein, "/" indicates adjacent lamination of layers. Hereinafter, the same).

Herein, the light emitting layer is a layer having function to emit a light, the hole transporting layer is a layer having function to transport a hole, and the electron transporting layer is a layer having function to transport an electron. Herein, the electron transporting layer and the hole transporting layer are generically called a charge transporting layer.

The light emitting layer, hole transporting layer and electron transporting layer also may be used each independently in two or more layers.

Of charge transporting layers disposed adjacent to an electrode, that having function to improve charge injecting efficiency from the electrode and having effect to decrease driving voltage of an device are particularly called sometimes a charge injecting layer (hole injecting layer, electron

injecting layer) in general.

For enhancing adherence with an electrode and improving charge injection from an electrode, the above-described charge injecting layer or insulation layer having a thickness of 2 nm or less may also be provided adjacent to an electrode, and further, for enhancing adherence of the interface, preventing mixing and the like, a thin buffer layer may also be inserted into the interface of a charge transporting layer and light emitting layer.

The order and number of layers laminated and the thickness of each layer can be appropriately applied while considering light emitting efficiency and life of the device.

In the present invention, as the polymer LED having a charge injecting layer (electron injecting layer, hole injecting layer) provided, there are listed a polymer LED having a charge injecting layer provided adjacent to a cathode and a polymer LED having a charge injecting layer provided adjacent to an anode.

For example, the following structures e) to p) are specifically exemplified.

e) anode/charge injecting layer/light emitting layer/cathode

f) anode/light emitting layer/charge injecting layer/cathode

g) anode/charge injecting layer/light emitting layer/charge injecting layer/cathode

h) anode/charge injecting layer/hole transporting layer/light emitting layer/cathode

i) anode/hole transporting layer/light emitting layer/charge injecting layer/cathode

j) anode/charge injecting layer/hole transporting layer/light emitting layer/charge injecting layer/cathode

k) anode/charge injecting layer/light emitting layer/electron transporting layer/cathode

l) anode/light emitting layer/electron transporting layer/charge injecting layer/cathode



m) anode/charge injecting layer/light emitting layer/electron transporting layer/charge injecting layer/cathode

n) anode/charge injecting layer/hole transporting layer/light emitting layer/electron transporting layer/cathode

o) anode/hole transporting layer/light emitting layer/electron transporting layer/charge injecting layer/cathode

p) anode/charge injecting layer/hole transporting layer/light emitting layer/electron transporting layer/charge injecting layer/cathode

As the specific examples of the charge injecting layer, there are exemplified layers containing an conducting polymer, layers which are disposed between an anode and a hole transporting layer and contain a material having an ionization potential between the ionization potential of an anode material and the ionization potential of a hole transporting material contained in the hole transporting layer, layers which are disposed between a cathode and an electron transporting layer and contain a material having an electron affinity between the electron affinity of a cathode material and the electron affinity of an electron transporting material contained in the electron transporting layer, and the like.

When the above-described charge injecting layer is a layer containing an conducting polymer, the electric conductivity of the conducting polymer is preferably  $10^{-5}$  S/cm or more and  $10^3$  S/cm or less, and for decreasing the leak current between light emitting pixels, more preferably  $10^{-5}$  S/cm or more and  $10^2$  S/cm or less, further preferably  $10^{-5}$  S/cm or more and  $10^1$  S/cm or less.

Usually, to provide an electric conductivity of the conducting polymer of  $10^{-5}$  S/cm or more and  $10^3$  S/cm or less, a suitable amount of ions are doped into the conducting polymer.

Regarding the kind of an ion doped, an anion is used in

a hole injecting layer and a cation is used in an electron injecting layer. As examples of the anion, a polystyrene sulfonate ion, alkylbenzene sulfonate ion, camphor sulfonate ion and the like are exemplified, and as examples of the cation, a lithium ion, sodium ion, potassium ion, tetrabutyl ammonium ion and the like are exemplified.

The thickness of the charge injecting layer is for example, from 1 nm to 100 nm, preferably from 2 nm to 50 nm.

Materials used in the charge injecting layer may properly be selected in view of relation with the materials of electrode and adjacent layers, and there are exemplified conducting polymers such as polyaniline and derivatives thereof, polythiophene and derivatives thereof, polypyrrole and derivatives thereof, poly(phenylene vinylene) and derivatives thereof, poly(thienylene vinylene) and derivatives thereof, polyquinoline and derivatives thereof, polyquinoxaline and derivatives thereof, polymers containing aromatic amine structures in the main chain or the side chain, and the like, and metal phthalocyanine (copper phthalocyanine and the like), carbon and the like.

The insulation layer having a thickness of 2 nm or less has function to make charge injection easy. As the material of the above-described insulation layer, metal fluoride, metal oxide, organic insulation materials and the like are listed. As the polymer LED having an insulation layer having a thickness of 2 nm or less, there are listed polymer LEDs having an insulation layer having a thickness of 2 nm or less provided adjacent to a cathode, and polymer LEDs having an insulation layer having a thickness of 2 nm or less provided adjacent to an anode.

[0196]

Specifically, there are listed the following structures q) to ab) for example.

q) anode/insulation layer having a thickness of 2 nm or less/light emitting layer/cathode

r) anode/light emitting layer/insulation layer having a

thickness of 2 nm or less/cathode

s) anode/insulation layer having a thickness of 2 nm or less/light emitting layer/insulation layer having a thickness of 2 nm or less/cathode

t) anode/insulation layer having a thickness of 2 nm or less/hole transporting layer/light emitting layer/cathode

u) anode/hole transporting layer/light emitting layer/insulation layer having a thickness of 2 nm or less/cathode

v) anode/insulation layer having a thickness of 2 nm or less/hole transporting layer/light emitting layer/insulation layer having a thickness of 2 nm or less/cathode

w) anode/insulation layer having a thickness of 2 nm or less/light emitting layer/electron transporting layer/cathode

x) anode/light emitting layer/electron transporting layer/insulation layer having a thickness of 2 nm or less/cathode

y) anode/insulation layer having a thickness of 2 nm or less/light emitting layer/electron transporting layer/insulation layer having a thickness of 2 nm or less/cathode

z) anode/insulation layer having a thickness of 2 nm or less/hole transporting layer/light emitting layer/electron transporting layer/cathode

aa) anode/hole transporting layer/light emitting layer/electron transporting layer/insulation layer having a thickness of 2 nm or less/cathode

ab) anode/insulation layer having a thickness of 2 nm or less/hole transporting layer/light emitting layer/electron transporting layer/insulation layer having a thickness of 2 nm or less/cathode

[0197]

Regarding the thickness of the light emitting layer, the optimum value differs depending on material used, and may properly be selected so that the driving voltage and the light

emitting efficiency become optimum values, and for example, it is from 1 nm to 1  $\mu$ m, preferably from 2 nm to 500 nm, further preferably from 5 nm to 200 nm.

In the polymer LED of the present invention, light emitting materials other than the above-described polymeric fluorescent substance can also be mixed in a light emitting layer. Further, in the polymer LED of the present invention, the light emitting layer containing light emitting materials other than the above-described polymeric fluorescent substance may also be laminated with a light emitting layer containing the above-described polymeric fluorescent substance.

As the light emitting material, known materials can be used. In a compound having lower molecular weight, there can be used, for example, naphthalene derivatives, anthracene or derivatives thereof, perylene or derivatives thereof; dyes such as polymethine dyes, xanthene dyes, coumarine dyes, cyanine dyes; metal complexes of 8-hydroxyquinoline or derivatives thereof, aromatic amine, tetraphenylcyclopentane or derivatives thereof, or tetraphenylbutadiene or derivatives thereof, and the like.

Specifically, there can be used known compounds such as those described in JP-A Nos. 57-51781, 59-195393 and the like, for example.

When the polymer LED of the present invention has a hole transporting layer, as the hole transporting materials used, there are exemplified polyvinylcarbazole or derivatives thereof, polysilane or derivatives thereof, polysiloxane derivatives having an aromatic amine in the side chain or the main chain, pyrazoline derivatives, arylamine derivatives, stilbene derivatives, triphenyldiamine derivatives, polyaniline or derivatives thereof, polythiophene or derivatives thereof, polypyrrole or derivatives thereof, poly(p-phenylenevinylene) or derivatives thereof, poly(2,5-thienylenevinylene) or derivatives thereof, or the like.

Specific examples of the hole transporting material include those described in JP-A Nos. 63-70257, 63-175860, 2-135359, 2-135361, 2-209988, 3-37992 and 3-152184.

Among them, as the hole transporting materials used in the hole transporting layer, preferable are polymer hole transporting materials such as polyvinylcarbazole or derivatives thereof, polysilane or derivatives thereof, polysiloxane derivatives having an aromatic amine compound group in the side chain or the main chain, polyaniline or derivatives thereof, polythiophene or derivatives thereof, poly(p-phenylenevinylene) or derivatives thereof, poly(2,5-thienylenevinylene) or derivatives thereof, or the like, and further preferable are polyvinylcarbazole or derivatives thereof, polysilane or derivatives thereof and polysiloxane derivatives having an aromatic amine compound group in the side chain or the main chain. In the case of a hole transporting material having lower molecular weight, it is preferably dispersed in a polymer binder for use.

Polyvinylcarbazole or derivatives thereof are obtained, for example, by cation polymerization or radical polymerization from a vinyl monomer.

As the polysilane or derivatives thereof, there are exemplified compounds described in Chem. Rev., 89, 1359 (1989) and GB 2300196 published specification, and the like. For synthesis, methods described in them can be used, and a Kipping method can be suitably used particularly.

As the polysiloxane or derivatives thereof, those having the structure of the above-described hole transporting material having lower molecular weight in the side chain or main chain, since the siloxane skeleton structure has poor hole transporting property. Particularly, there are exemplified those having an aromatic amine having hole transporting property in the side chain or main chain.

The method for forming a hole transporting layer is not restricted, and in the case of a hole transporting layer having lower molecular weight, a method in which the layer is formed

from a mixed solution with a polymer binder is exemplified. In the case of a polymer hole transporting material, a method in which the layer is formed from a solution is exemplified.

The solvent used for the film forming from a solution is not particularly restricted providing it can dissolve a hole transporting material. As the solvent, there are exemplified chlorine solvents such as chloroform, methylene chloride, dichloroethane and the like, ether solvents such as tetrahydrofuran and the like, aromatic hydrocarbon solvents such as toluene, xylene and the like, ketone solvents such as acetone, methyl ethyl ketone and the like, and ester solvents such as ethyl acetate, butyl acetate, ethylcellosolve acetate and the like.

As the film forming method from a solution, there can be used coating methods such as a spin coating method, casting method, micro gravure coating method, gravure coating method, bar coating method, roll coating method, wire bar coating method, dip coating method, spray coating method, screen printing method, flexo printing method, offset printing method, inkjet printing method and the like, from a solution.

The polymer binder mixed is preferably that does not disturb charge transport extremely, and that does not have strong absorption of a visible light is suitably used. As such polymer binder, polycarbonate, polyacrylate, poly(methyl acrylate), poly(methyl methacrylate), polystyrene, poly(vinyl chloride), polysiloxane and the like are exemplified.

[0219]

Regarding the thickness of the hole transporting layer, the optimum value differs depending on material used, and may properly be selected so that the driving voltage and the light emitting efficiency become optimum values, and at least a thickness at which no pin hole is produced is necessary, and too large thickness is not preferable since the driving voltage of the device increases. Therefore, the thickness of the hole transporting layer is, for example, from 1 nm to 1  $\mu$  m,

preferably from 2 nm to 500 nm, further preferably from 5 nm to 200 nm.

[0220]

The substrate forming the polymer LED of the present invention may preferably be that does not change in forming an electrode and layers of organic materials, and there are exemplified glass, plastics, polymer film, silicon substrates and the like. In the case of a opaque substrate, it is preferable that the opposite electrode is transparent or semitransparent.

[0221]

In the present invention, it is preferable that the anode is transparent or semitransparent.

As the material of this anode, electron conductive metal oxide films, semitransparent metal thin films and the like are used. Specifically, there are used indium oxide, zinc oxide, tin oxide, and films (NESA and the like) fabricated by using an electron conductive glass composed of indium/tin/oxide (ITO), indium/zinc/oxide and the like, which are metal oxide complexes, and gold, platinum, silver, copper and the like are used, and among them, ITO, indium/zinc/oxide, tin oxide are preferable. As the fabricating method, a vacuum vapor deposition method, sputtering method, ion plating method, plating method and the like are used. As the anode, there may also be used organic transparent conducting films such as polyaniline or derivatives thereof, polythiophene or derivatives thereof and the like.

[0222]

The thickness of the anode can be appropriately selected while considering transmission of a light and electric conductivity, and for example, from 10 nm to 10  $\mu\text{m}$ , preferably from 20 nm to 1  $\mu\text{m}$ , further preferably from 50 nm to 500 nm.

[0223]

Further, for easy charge injection, there may be provided on the anode a layer comprising a phthalocyanine derivative conducting polymers, carbon and the like, or a layer having

an average film thickness of 2 nm or less comprising a metal oxide, metal fluoride, organic insulating material and the like.

[0224]

As the material of a cathode used in the polymer LED of the present invention, that having lower work function is preferable. For example, there are used metals such as lithium, sodium, potassium, rubidium, cesium, beryllium, magnesium, calcium, strontium, barium, aluminum, scandium, vanadium, zinc, yttrium, indium, cerium, samarium, europium, terbium, ytterbium and the like, or alloys comprising two or more of them, or alloys comprising one or more of them with one or more of gold, silver, platinum, copper, manganese, titanium, cobalt, nickel, tungsten and tin, graphite or graphite intercalation compounds and the like. Examples of alloys include a magnesium-silver alloy, magnesium-indium alloy, magnesium-aluminum alloy, indium-silver alloy, lithium-aluminum alloy, lithium-magnesium alloy, lithium-indium alloy, calcium-aluminum alloy and the like. The cathode may be formed into a laminated structure of two or more layers.

[0225]

The thickness of the cathode can be appropriately selected while considering transmission of a light and electric conductivity, and for example, from 10 nm to 10  $\mu$  m, preferably from 20 nm to 1  $\mu$  m, further preferably from 50 nm to 500 nm.

[0226]

As the method for fabricating a cathode, there are used a vacuum vapor deposition method, sputtering method, lamination method in which a metal thin film is adhered under heat and pressure, and the like. Further, there may also be provided, between a cathode and an organic layer, a layer comprising an conducting polymer, or a layer having an average film thickness of 2 nm or less comprising a metal oxide, metal fluoride, organic insulation material and the like, and after



fabrication of the cathode, a protective layer may also be provided which protects the polymer LED. For stable use of the polymer LED for a long period of time, it is preferable to provide a protective layer and/or protective cover for protection of the device in order to prevent it from outside damage.

[0227]

As the protective layer, there can be used a polymeric compound, metal oxide, metal fluoride, metal borate and the like. As the protective cover, there can be used a glass plate, a plastic plate the surface of which has been subjected to lower-water-permeation treatment, and the like, and there is suitably used a method in which the cover is pasted with an device substrate by a thermosetting resin or light-curing resin for sealing. If space is maintained using a spacer, it is easy to prevent an device from being injured. If an inner gas such as nitrogen and argon is sealed in this space, it is possible to prevent oxidation of a cathode, and further, by placing a desiccant such as barium oxide and the like in the above-described space, it is easy to suppress the damage of an device by moisture adhered in the production process. Among them, any one means or more are preferably adopted.

[0228]

The polymer LED of the present invention can be used for a flat light source, a segment display, a dot matrix display, and a liquid crystal display as a back light, etc.

For obtaining light emission in plane form using the polymer LED of the present invention, an anode and a cathode in the plane form may properly be placed so that they are laminated each other. Further, for obtaining light emission in pattern form, there is a method in which a mask with a window in pattern form is placed on the above-described plane light emitting device, a method in which an organic layer in non-light emission part is formed to obtain extremely large thickness providing substantial non-light emission, and a method in which any one of an anode or a cathode, or both of them are

formed in the pattern. By forming a pattern by any of these methods and by placing some electrodes so that independent on/off is possible, there is obtained a display device of segment type which can display digits, letters, simple marks and the like. Further, for forming a dot matrix device, it may be advantageous that anodes and cathodes are made in the form of stripes and placed so that they cross at right angles. By a method in which a plurality of kinds of polymeric compounds emitting different colors of lights are placed separately or a method in which a color filter or luminescence converting filter is used, area color displays and multi color displays are obtained. A dot matrix display can be driven by passive driving, or by active driving combined with TFT and the like. These display devices can be used as a display of a computer, television, portable terminal, portable telephone, car navigation, view finder of a video camera, and the like.

[0229]

Further, the above-described light emitting device in plane form is a thin self-light-emitting one, and can be suitably used as a flat light source for back-light of a liquid crystal display, or as a flat light source for illumination. Further, if a flexible plate is used, it can also be used as a curved light source or a display.

[0230]

[Effect of the Invention]

The complex and the polymer compound of the present invention have phosphorescence in a visible region, and are excellent in light emitting efficiency.

[Name of Document] ABSTRACT

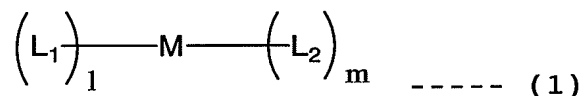
[Abstract]

[Problem]

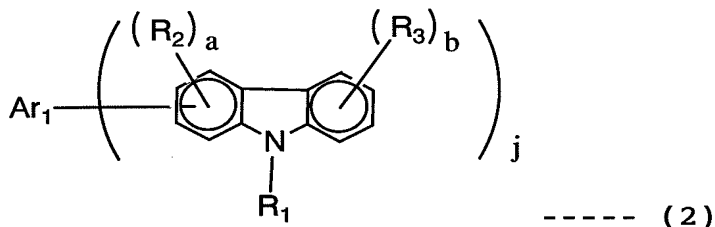
To provide a complex having good light emitting efficiency and phosphorescence in visible region.

[Solving Means]

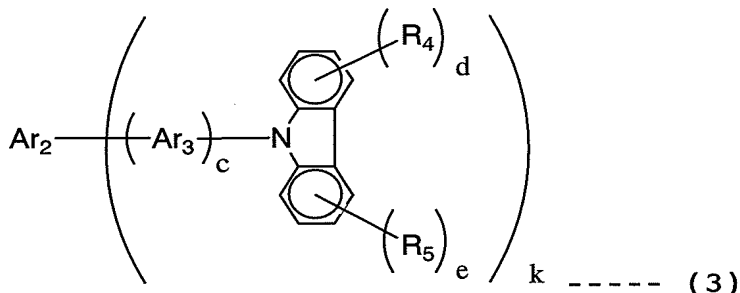
A complex represented by formula (1) and having phosphorescence in a visible region.



[M is a metal.  $L_1$  is a ligand represented by formula (2) or (3).  $L_2$  is a ligand which bonds with M by nitrogen atom etc. 1 is an integer of 1-3. 1+m is an integer of 2-5.]



( $Ar_1$  is a ligand having a substituent of carbazole ring structure, which bonds to M by nitrogen atom, etc., and has j pieces of substituents having carbazole ring structure. j represents an integer of 1 to 3. j represents an integer of 1 to 3. a is an integer of 0-3. b is an integer of 0-4.)



( $Ar_2$  is a ligand which bonds to M by nitrogen atom, etc., and has covalent bonds to k pieces of  $Ar_2$ s. k represents an integer of 1-3. k is an integer of 1-3.  $Ar_3$  is an alkylene group etc. k is an integer of 1-3. c is 0 or 1. d and e are integers of

0-3 ).

[Selected Figure] No